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Müller et al.

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[54] **HERBICIDAL SUBSTITUTED
4-SULPHONYLAMINO-2-AZINYL-1,2,4-
TRIAZOL-3-ONES**

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[21] **Appl. No.:** 583,915

[22] **Filed:** Sep. 17, 1990

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4,988,381.

Foreign Application Priority Data

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Jan. 16, 1989 [DE] Fed. Rep. of Germany 3901084

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C07D 403/04; C07D 403/14

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544/207; 544/209; 544/212

[58] **Field of Search** 71/93; 544/198, 207,
544/209, 212

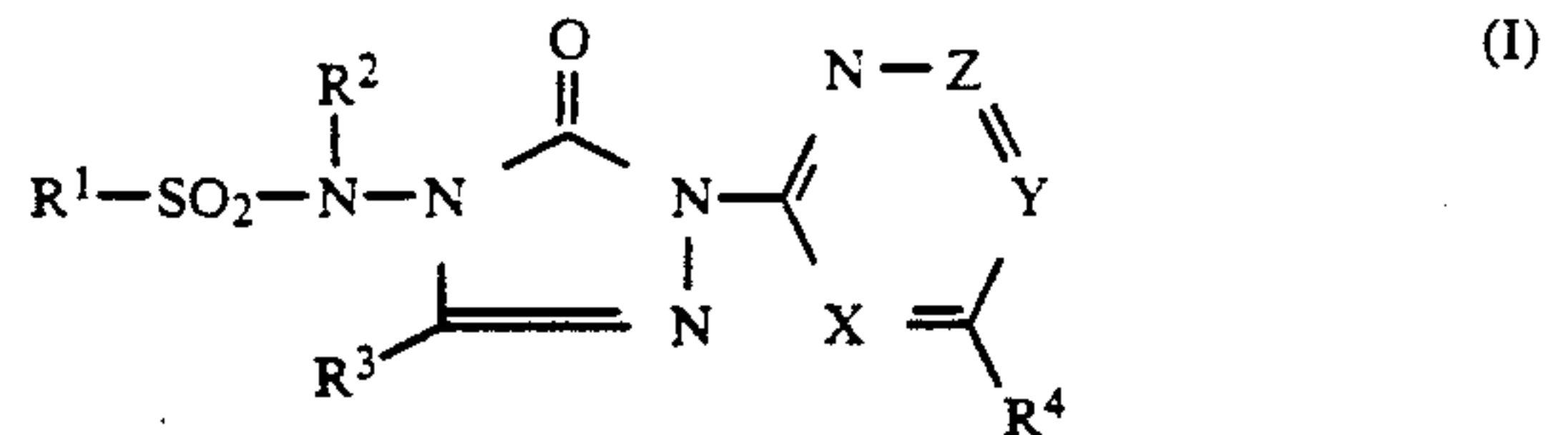
References Cited**U.S. PATENT DOCUMENTS**

4,988,811 1/1991 Valbusa et al. 544/198

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Woods

ABSTRACT

Herbicide substituted sulphonylamino triazolinones of
the formula



in which

R¹ represents an optionally substituted radical from
the group consisting of alkyl, aralkyl, aryl and
heteroaryl,

R² represents hydrogen or the group —SO₂—R¹
where R¹ has the abovementioned meaning,

R³ represents hydrogen, halogen, hydroxyl, mer-
capto, amino or an optionally substituted radical
from the group consisting of alkyl, cycloalkyl,
aralkyl, aryl, alkoxy, alkenyloxy, alkinyloxy, alkyl-
thio, alkylsulphinyl, alkylsulphonyl, alkenylthio,
alkinylthio, aralkoxy, aralkylthio, alkylamino and
dialkylamino,

R⁴ represents hydrogen, halogen, hydroxyl, amino or
an optionally substituted radical from the group
consisting of alkyl, alkoxy, alkylthio, alkylsulphi-
nyl, alkylsulphonyl, alkylamino and dialkylamino,

X represents nitrogen or a CH group,

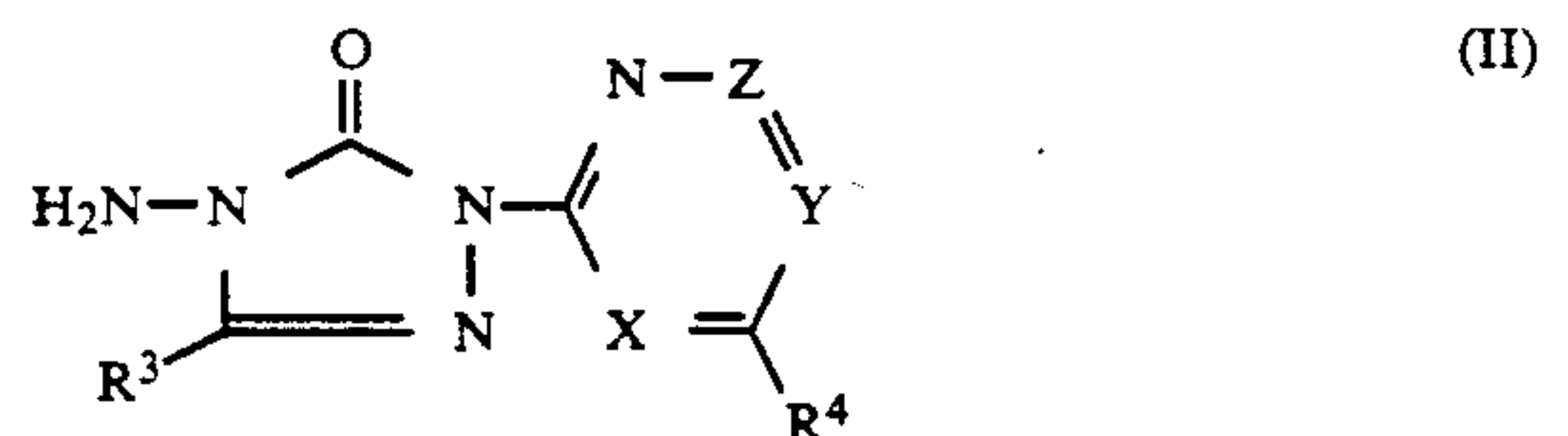
Y represents nitrogen or a CR⁵ group where

R⁵ represents hydrogen, halogen, cyano, alkyl,
formyl, alkylcarbonyl or alkoxy carbonyl, and

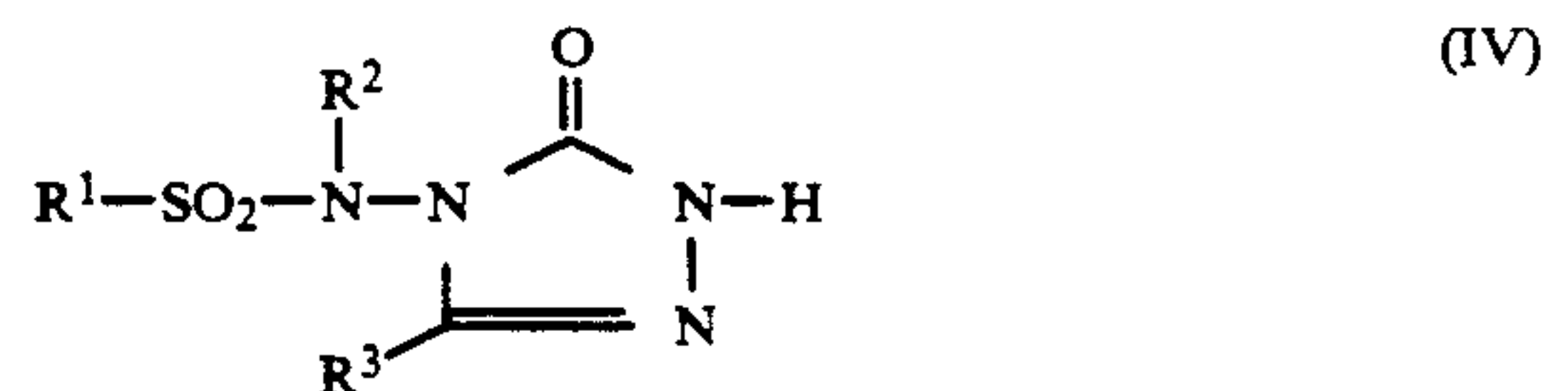
Z represents nitrogen or a CR⁶ group where

R⁶ represents hydrogen, halogen, hydroxyl, amino
or an optionally substituted radical from the
group consisting of alkyl, alkoxy, alkylthio, al-
kylsulphinyl, alkylsulphonyl, alkylamino and
dialkylamino,

and salts thereof. Intermediates of the formulas



and



are also new.

4 Claims, No Drawings

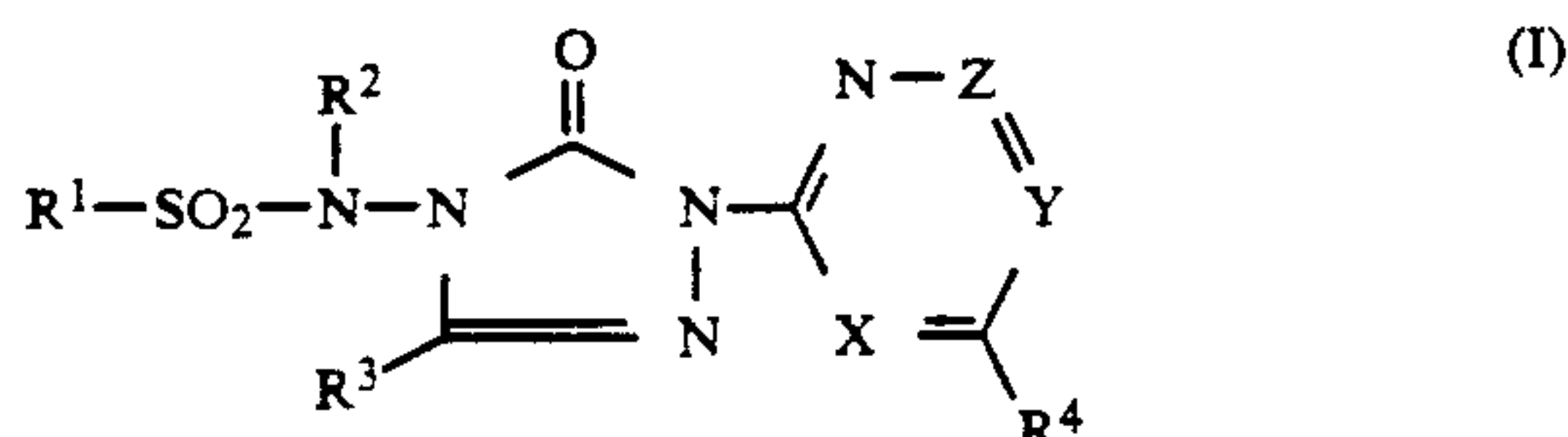
**HERBICIDAL SUBSTITUTED
4-SULPHONYLAMINO-2-AZINYL-1,2,4-TRIAZOL-
3-ONES**

This is a division of application Ser. No. 382,163, filed July 19, 1989, now issued U.S. Pat. No. 4,988,381.

The invention relates to new substituted 4-sulphonylamino-2-azinyl-1,2,4-triazol-3-ones (hereinafter abbreviated to "substituted sulphonylamino-1,2,4-triazol-3-ones"), processes and new intermediates for their preparation, and their use as herbicides.

It has been disclosed that certain substituted triazolones, such as, for example, 4-(3-trifluoromethoxybenzylideneamino)-2,4-dihydro-3H-1,2,4-triazol-3-one, are herbicidally active (cf. U.S. Pat. No. 3,884,910). However, the action of these known compounds is not satisfactory in all respects.

The new substituted sulphonylamino-1,2,4-triazolones of the general formula (I)



in which

R¹ represents an optionally substituted radical from the series comprising alkyl, aralkyl, aryl and heteroaryl,

R² represents hydrogen or the group -SO₂-R¹ where

R¹ has the abovementioned meaning,

R³ represents hydrogen, halogen, hydroxyl, mercapto, amino or an optionally substituted radical from the series comprising alkyl, cycloalkyl, aralkyl, aryl, alkoxy, alkenyloxy, alkinyl, alkythio, alkylsulphinyl, alkylsulphonyl, alkenylthio, alkinylthio, aralkoxy, aralkylthio, alkylamino and dialkylamino,

R⁴ represents hydrogen, halogen, hydroxyl, amino or an optionally substituted radical from the series comprising alkyl, alkoxy, alkylthio, alkylsulphinyl, alkylsulphonyl, alkylamino and dialkylamino,

X represents nitrogen or a CH group,

Y represents nitrogen or a CR⁵ group where

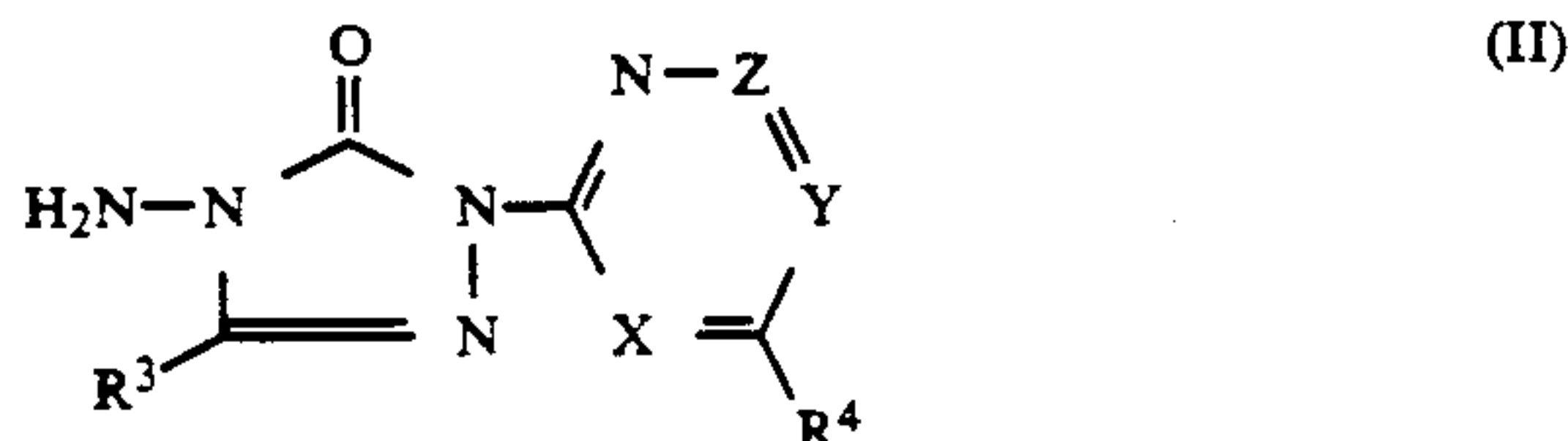
R⁵ represents hydrogen, halogen, cyano, alkyl, formyl, alkylcarbonyl or alkoxy-carbonyl, and

Z represents nitrogen or a CR⁶ group where

R⁶ represents hydrogen, halogen, hydroxyl, amino or an optionally substituted radical from the series comprising alkyl, alkoxy, alkylthio, alkylsulphinyl, alkylsulphonyl, alkylamino and dialkylamino, and salts of compounds of the formula (I) have now been found.

The new substituted sulphonylamino-1,2,4-triazolones of the general formula (I) are obtained when

(a) substituted aminotriazolones of the general formula (II)



in which

R³, R⁴, X, Y and Z have the abovementioned meanings,

are reacted with sulphonyl halides or sulphonic anhydrides of the general formula (III)



in which

R¹ has the abovementioned meaning and

Q represents fluorine, chlorine, bromine or the group -O-SO₂-R¹, where

R¹ has the abovementioned meaning,

if appropriate, in the presence of an acid acceptor and, if appropriate, in the presence of a diluent, the resulting compounds of the formula (I) in which

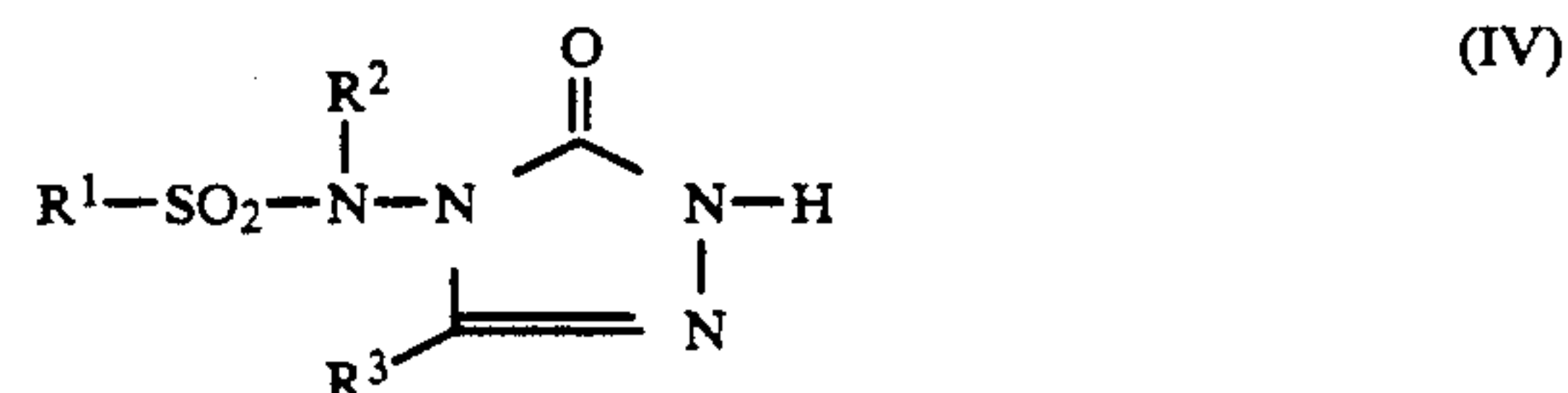
R² represents the group -SO₂-R¹,

are reacted, if appropriate, with desulphonylating agents, if appropriate in the presence of diluents, to give compounds of the formula (I) in which

R² represents hydrogen,

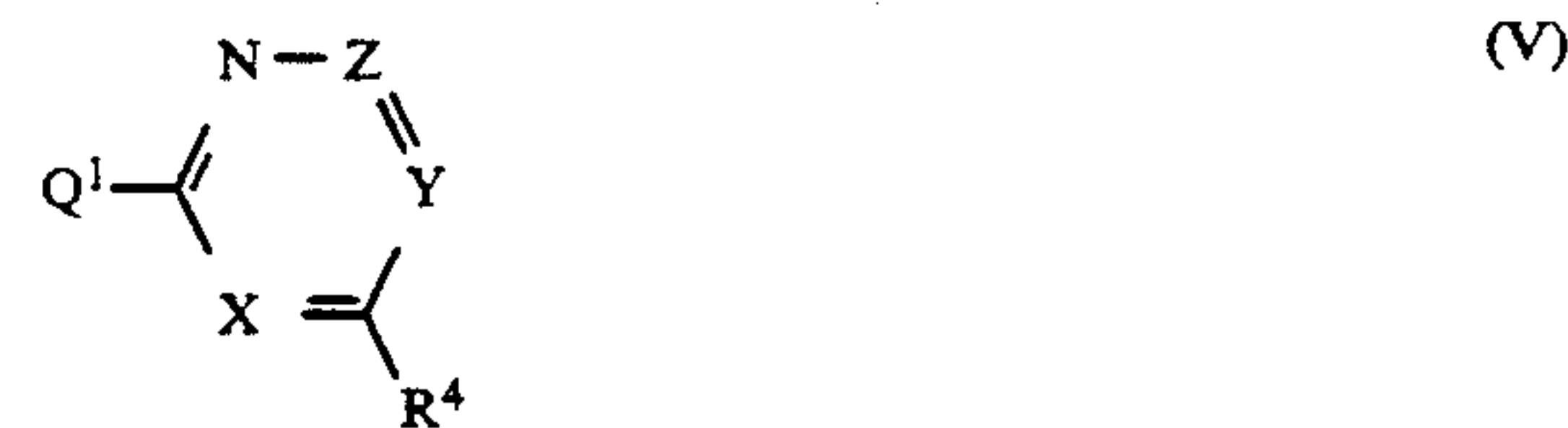
and, if appropriate, salts are prepared therefrom by customary methods or when

b) sulphonylamino-1,2,4-triazolones of the general formula (IV)



in which

R¹, R² and R³ have the above-mentioned meanings, are reacted with azines of the general formula (V)



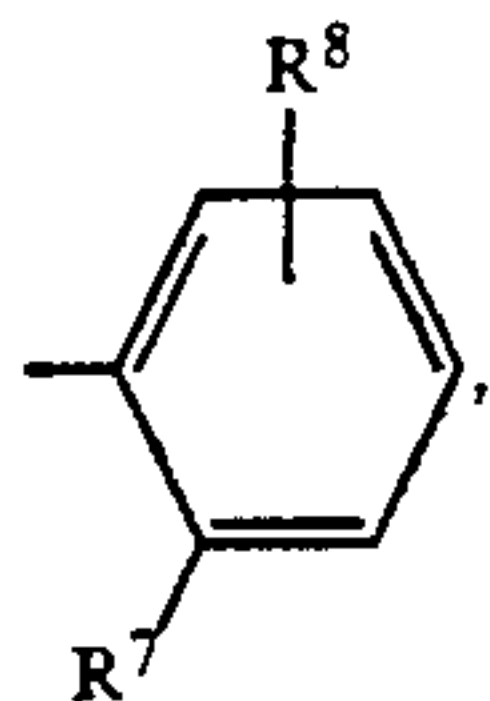
in which

R⁴, X, Y and Z have the above-mentioned meanings and

Q¹ represents halogen, benzylsulphonyl or alkylsulphonyl, if appropriate, in the presence of an acid acceptor and, if appropriate, in the presence of a diluent and, if appropriate, salts are prepared by customary methods from the resultant compounds of the formula (I).

The new substituted sulphonylamino-1,2,4-triazolones of the general formula (I) are distinguished by powerful herbicidal activity. These compounds represent a chemically novel class of herbicides. Surprisingly, the new compounds of the formula (I) show a considerably better herbicidal action than 4-(3-trifluoromethoxybenzylideneamino)-2,4-dihydro-3H-1,2,4-triazol-3-one, which is known and has similar structure.

The invention preferably relates to compounds of the formula (I) in which R¹ represents the radical



where

R⁷ and R⁸ are identical or different and represent hydrogen, fluorine, chlorine, bromine, iodine, cyano, nitro, C₁-C₆-alkyl [which is optionally substituted by fluorine, chlorine, bromine, cyano, carboxyl, C₁-C₄-alkoxycarbonyl, C₁-C₄-alkylaminocarbonyl, di-(C₁-C₄-alkyl)aminocarbonyl, hydroxyl, C₁-C₄-alkoxy, formyloxy, C₁-C₄-alkyl-carbonyloxy, C₁-C₄-alkoxycarbonyloxy, C₁-C₄-alkylamino-carbonyloxy, C₁-C₄-alkylthio, C₁-C₄-alkylsulphinyl, C₁-C₄-alkylsulphonyl, di-(C₁-C₄-alkyl)aminosulphonyl, C₃-C₆-cycloalkyl or phenyl], represent C₂-C₆-alkenyl [which is optionally substituted by fluorine, chlorine, bromine, cyano, C₁-C₄-alkoxycarbonyl, carboxyl or phenyl], represent C₂-C₆-alkinyl [which is optionally substituted by fluorine, chlorine, bromine, cyano, C₁-C₄-alkoxy-carbonyl, carboxyl or phenyl], represent C₁-C₄-alkoxy [which is optionally substituted by fluorine, chlorine, bromine, cyano, carboxyl, C₁-C₄-alkoxy-carbonyl, C₁-C₄-alkoxy, C₁-C₄-alkylthio, C₁-C₄-alkylsulphinyl or C₁-C₄-alkylsulphonyl], represent C₁-C₄-alkylthio [which is optionally substituted by fluorine, chlorine, bromine, cyano, carboxyl, C₁-C₄-alkoxy-carbonyl, C₁-C₄-alkylthio, C₁-C₄-alkylsulphinyl or C₁-C₄-alkylsulphonyl], represent C₃-C₆-alkenyloxy [which is optionally substituted by fluorine, chlorine, bromine, cyano or C₁-C₄-cyano or C₁-C₄-alkoxy-carbonyl], represent C₂-C₆-alkenylthio [which is optionally substituted by fluorine, chlorine, bromine, cyano, nitro, C₁-C₃-alkylthio or C₁-C₄-alkoxycarbonyl], C₃-C₆-alkinyloxy, C₃-C₆-alkinylthio or represent the radical -S(O)_p-R⁹, where

p represents the numbers 1 or 2 and

R⁹ represents fluorine, C₁-C₄-alkyl [which is optionally substituted by fluorine, chlorine, bromine, cyano or C₁-C₄-alkoxy-carbonyl], C₃-C₆-alkenyl, C₃-C₆-alkinyl, C₁-C₄-alkoxy, C₁-C₄-alkoxy-C₁-C₄-alkylamino, C₁-C₄-alkylamino, di-(C₁-C₄-alkyl)-amino or represents the radical -NHOR¹⁰, where

R¹⁰ represents C₁-C₆-alkyl [which is optionally substituted by fluorine, chlorine, cyano, C₁-C₄-alkoxy, C₁-C₄-alkylthio, C₁-C₄-alkylsulphinyl, C₁-C₄-alkylsulphonyl, C₁-C₄-alkyl-carbonyl, C₁-C₄-alkoxycarbonyl, C₁-C₄-alkylamino-carbonyl or di-(C₁-C₄-alkyl)aminocarbonyl], represents C₃-C₆-alkenyl [which is optionally substituted by fluorine, chlorine or bromine], C₃-C₆-alkinyl, C₃-C₆-cycloalkyl, C₃-C₆-cycloalkyl-C₁-C₂-alkyl, phenyl-C₁-C₂-alkyl [which is optionally substituted by fluorine, chlorine, nitro, cyano, C₁-C₄-alkyl, C₁-C₄-alkoxy or C₁-C₄-alkoxy-carbonyl], represents benzhydryl or represents phenyl [which is optionally substituted by fluorine, chlorine, nitro, cyano, C₁-C₄-alkyl, trifluoromethyl, C₁-C₄-alkoxy, C₁-C₂-fluoroalkoxy, C₁-C₄-alkylthio, trifluoromethylthio or C₁-C₄-alkoxycarbonyl],

R⁷ and/or R⁸ furthermore represent phenyl or phenoxy, C₁-C₄-alkylcarbonylamino, C₁-C₄-alkoxy-carbonylamino, C₁-C₄-alkylaminocarbonyl-amino, di-

(C₁-C₄-alkyl)-aminocarbonylamino, or represent the radical -CO-R¹¹, where

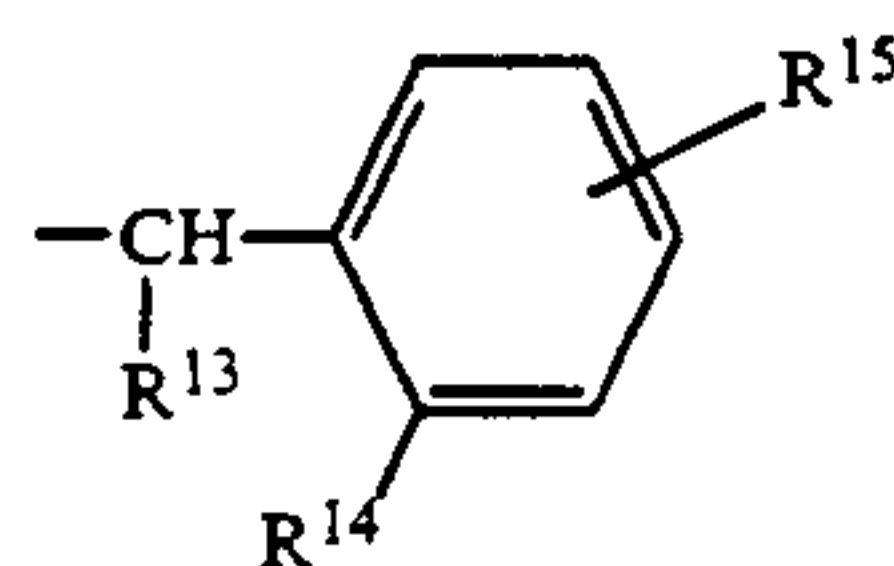
R¹¹ represents C₁-C₆-alkyl, C₁-C₆-alkoxy, C₃-C₆-cycloalkoxy, C₃-C₆-alkenyloxy, C₁-C₄ alkylthio, C₁-C₄-alkylamino, C₁-C₄-alkoxyamino, C₁-C₄-alkoxy-C₁-C₄-alkyl-amino or di-(C₁-C₄-alkyl)-amino [which are optionally substituted by fluorine and/or chlorine],

R⁷ and/or R⁸ furthermore represent C₁-C₄-alkylsulphonyloxy, di-(C₁-C₄-alkyl)-aminosulphonylamino or the radical -CH=N-R¹², where

R¹² represents C₁-C₆-alkyl which is optionally substituted by fluorine, chlorine, cyano, carboxyl, C₁-C₄-alkoxy, C₁-C₄-alkylthio, C₁-C₄-alkylsulphinyl or C₁-C₄-alkylsulphonyl, represents benzyl which is optionally substituted by fluorine or chlorine, represents C₃-C₆-alkenyl or C₃-C₆-alkinyl, each of which is optionally substituted by fluorine or chlorine, represents phenyl which is optionally substituted by fluorine, chlorine, bromine, C₁-C₄-alkyl, C₁-C₄-alkoxy, trifluoromethyl, trifluoromethoxy or trifluoromethylthio, represents C₁-C₆-alkoxy, C₃-C₆-alkenoxy, C₃-C₆-alkinoxy or benzyloxy, each of which is optionally substituted by fluorine and/or chlorine, represents amino, C₁-C₄-alkylamino, di-(C₁-C₄-alkyl)-amino, phenylamino, C₁-C₄-alkyl-carbonylamino, C₁-C₄-alkoxy-carbonylamino, C₁-C₄-alkylsulphonylamino or represents phenylsulphonylamino which is optionally substituted by fluorine, chlorine, bromine or methyl,

where furthermore

R¹ represents the radical

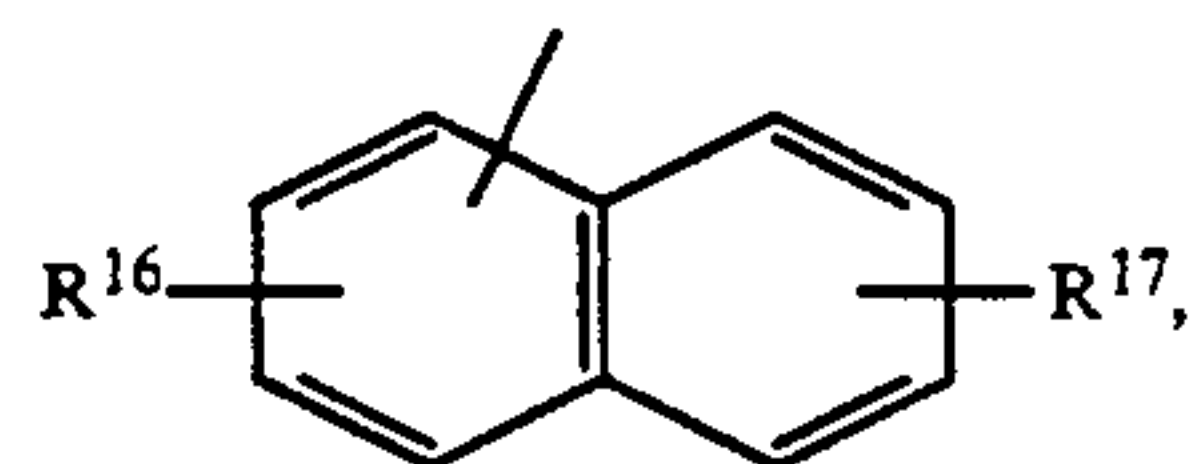


where

R¹³ represents hydrogen or C₁-C₄-alkyl,

R¹⁴ and R¹⁵ are identical or different and represent hydrogen, fluorine, chlorine, bromine, nitro, cyano, C₁-C₄-alkyl [which is optionally substituted by fluorine and/or chlorine], C₁-C₄-alkoxy [which is optionally substituted by fluorine and/or chlorine], carboxyl, C₁-C₄-alkoxycarbonyl, C₁-C₄-alkylsulphonyl or di-(C₁-C₄-alkyl)-aminosulphonyl; where furthermore

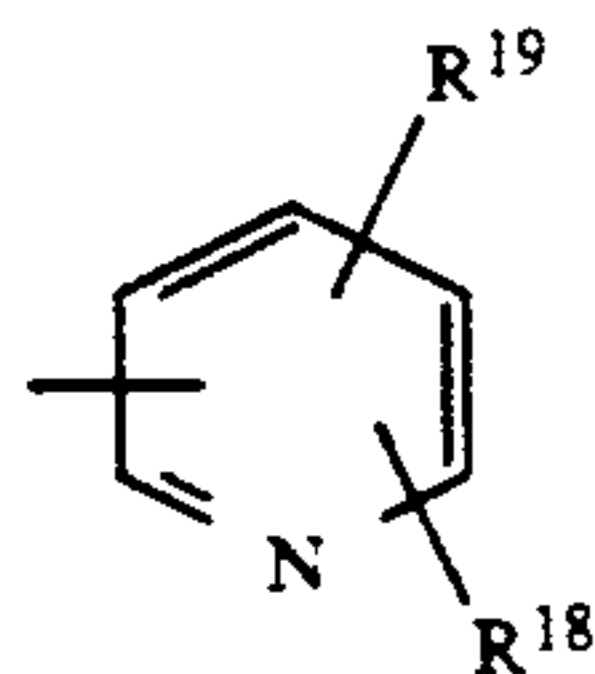
R¹ represents the radical



where

R¹⁶ and R¹⁷ are identical or different and represent hydrogen, fluorine, chlorine, bromine, nitro, cyano, C₁-C₄-alkyl [which is optionally substituted by fluorine and/or chlorine] or C₁-C₄-alkoxy [which is optionally substituted by fluorine and/or chlorine]; where furthermore

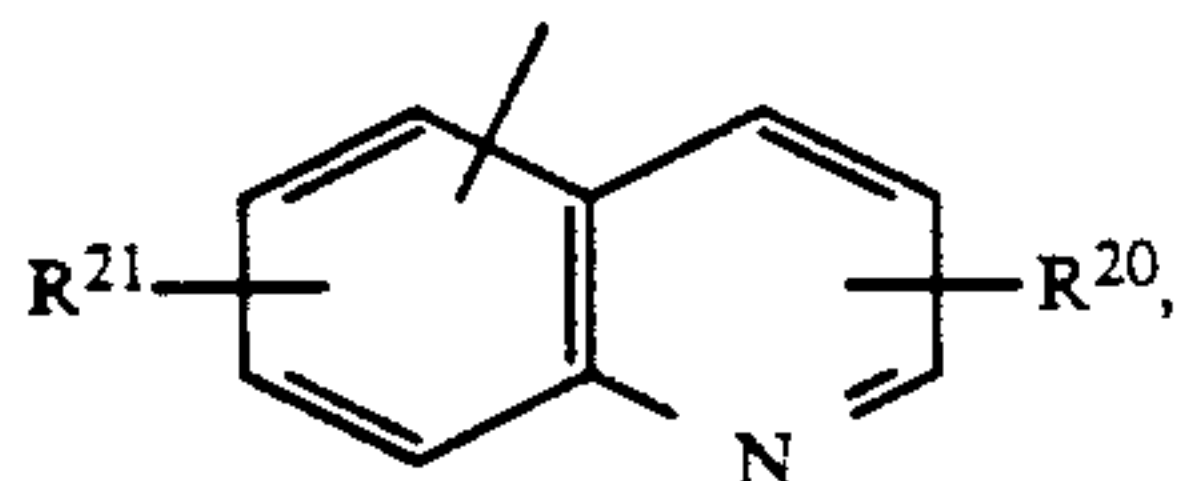
R¹ represents the radical



where

R¹⁸ and R¹⁹ are identical or different and represent hydrogen, fluorine, chlorine, bromine, nitro, cyano, C₁-C₄-alkyl [which is optionally substituted by fluorine and/or chlorine], C₂-C₄-alkenyl [which is optionally substituted by fluorine and/or chlorine], C₁-C₄-alkoxy [which is optionally substituted by fluorine and/or chlorine], represents C₁-C₄-alkylthio, C₁-C₄-alkylsulphinyl or C₁-C₄-alkylsulphonyl [which are optionally substituted by fluorine and/or chlorine], and represents di-(C₁-C₄-alkyl)-aminosulphonyl, C₁-C₄-alkoxy-carbonyl, dimethylaminocarbonyl or dioxolanyl; where furthermore

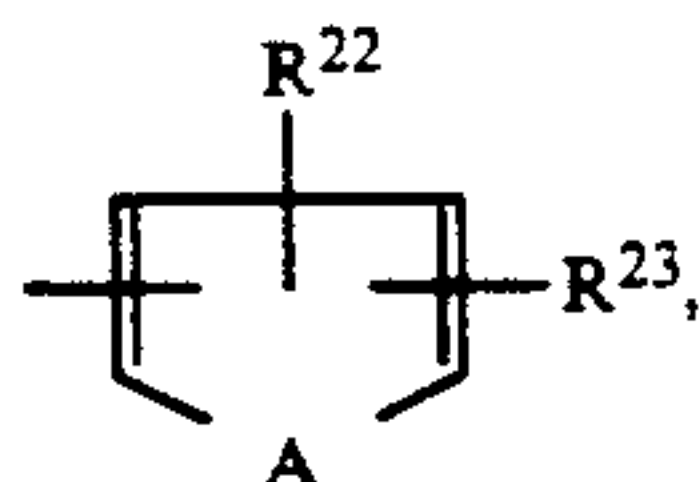
R¹ represents the radical



where

R²⁰ and R²¹ are identical or different and represent hydrogen, fluorine, chlorine, bromine, C₁-C₄-alkyl [which is optionally substituted by fluorine and/or bromine], C₁-C₄-alkoxy [which is optionally substituted by fluorine and/or chlorine], represent C₁-C₄-alkylthio, C₁-C₄-alkyl-sulphinyl or C₁-C₄-alkylsulphonyl [which are optionally substituted by fluorine and/or chlorine], or represent di-(C₁-C₄-alkyl)-aminosulphonyl; where furthermore

R¹ represents the radical



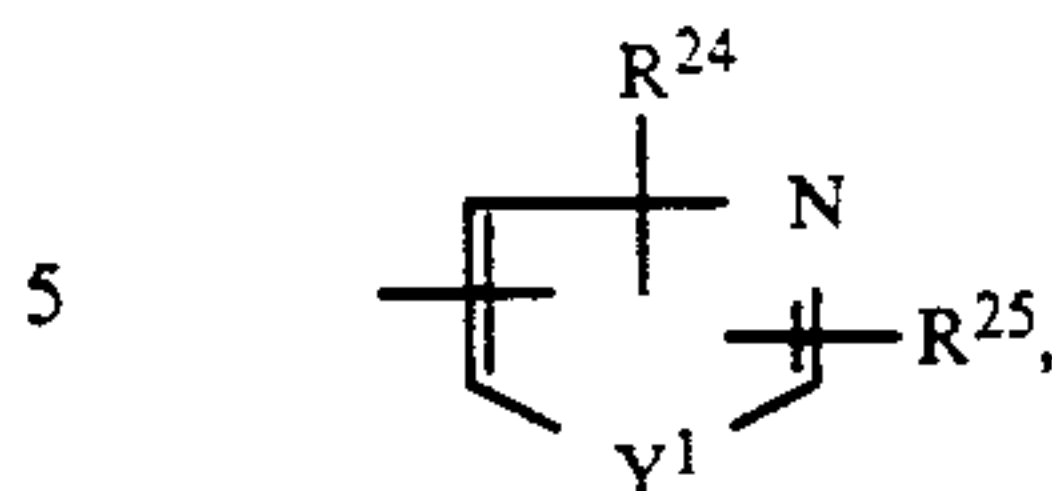
where

R²² and R²³ are identical or different and represent hydrogen, fluorine, chlorine, bromine, cyano, nitro, C₁-C₄-alkyl [which is optionally substituted by fluorine, chlorine, C₁-C₄-alkoxy and/or C₁-C₄-halogenoalkoxy], C₁-C₄-alkoxy [which is optionally substituted by fluorine and/or chlorine], C₁-C₁₄-alkylthio, C₁-C₄-alkylsulphinyl or C₁-C₄-alkylsulphonyl [which is optionally substituted by fluorine and/or chlorine], di-(C₁-C₄-alkyl)-aminosulphonyl or C₁-C₄-alkoxycarbonyl, and

A represents oxygen, sulphur or the group N—Z¹, where

Z¹ represents hydrogen, C₁-C₄-alkyl [which is optionally substituted by fluorine, chlorine, bromine or cyano], C₃-C₆-cycloalkyl, benzyl, phenyl [which is optionally substituted by fluorine, chlorine, bromine or nitro], C₁-C₄-alkylcarbonyl, C₁-C₄-alkoxy-carbonyl or di-(C₁-C₄-alkyl)-aminocarbonyl; where furthermore

R¹ represents the radical



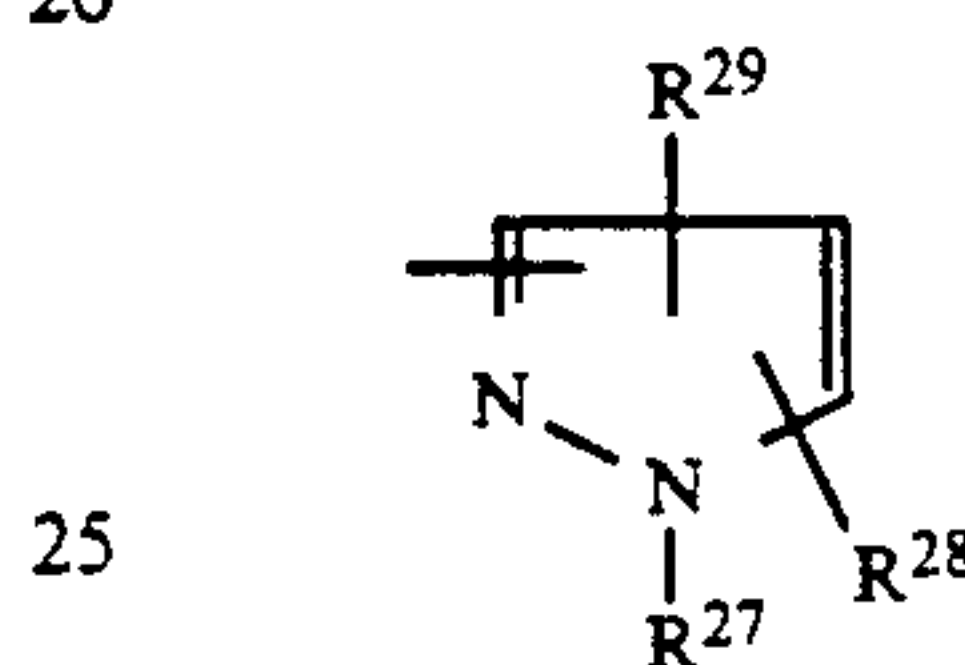
where

R²⁴ and R²⁵ are identical or different and represent hydrogen, C₁-C₄-alkyl, halogen, C₁-C₄-alkoxycarbonyl, C₁-C₄-alkoxy or C₁-C₄-halogenoalkoxy,

Y¹ represents sulphur or the group N—R²⁶, where

R²⁶ represents hydrogen or C₁-C₄-alkyl; where furthermore

R¹ represents the radical



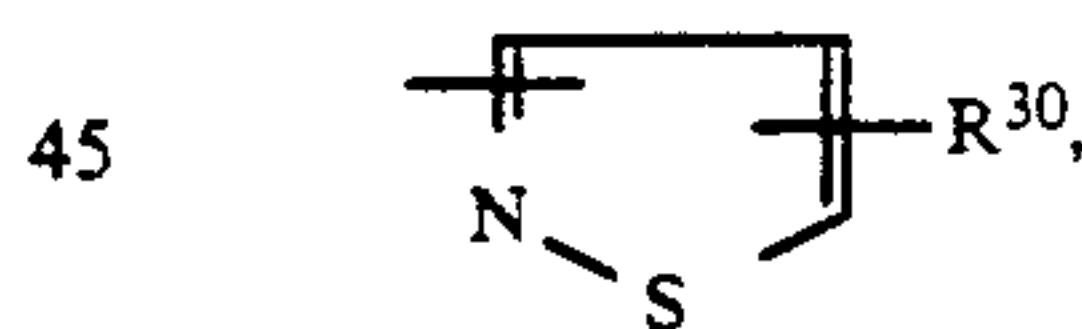
where

R²⁷ represents hydrogen, C₁-C₄-alkyl, benzyl, (iso)-quinolinylyl or phenyl,

R²⁸ represents hydrogen, halogen, cyano, nitro, C₁-C₄-alkyl [which is optionally substituted by fluorine and/or chlorine], C₁-C₄-alkoxy [which is optionally substituted by fluorine and/or chlorine], dioxolanyl or C₁-C₄-alkoxy-carbonyl and

R²⁹ represents hydrogen, halogen or C₁-C₄-alkyl; where furthermore

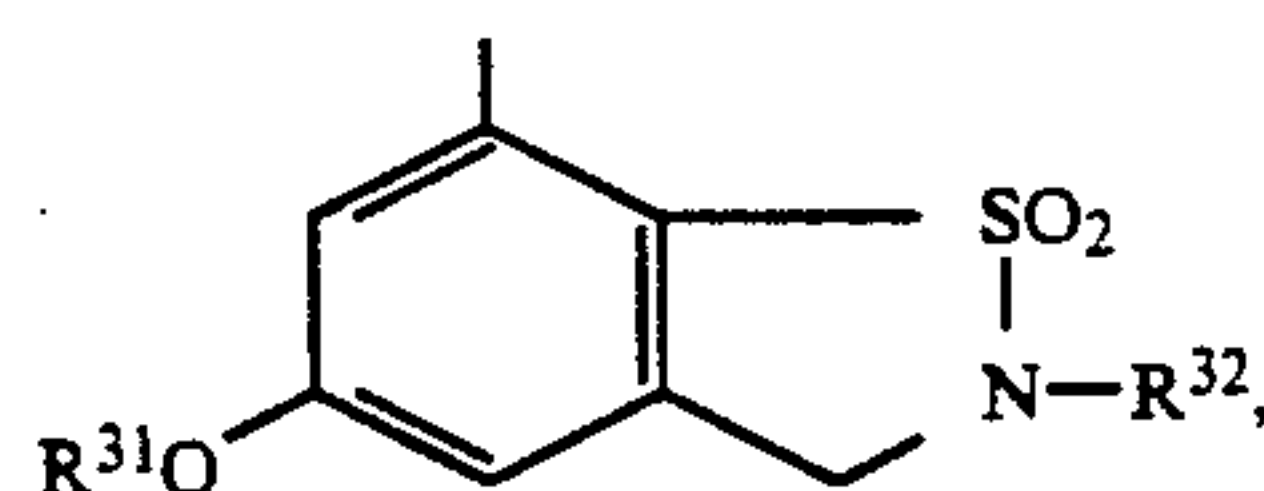
R¹ represents the radical



where

R³⁰ represents hydrogen, halogen, C₁-C₄-alkyl, C₁-C₄-halogenoalkyl, C₁-C₄-alkoxy, C₁-C₄-halogenoalkoxy or C₁-C₄-alkoxy-carbonyl; where furthermore

R¹ represents the radical

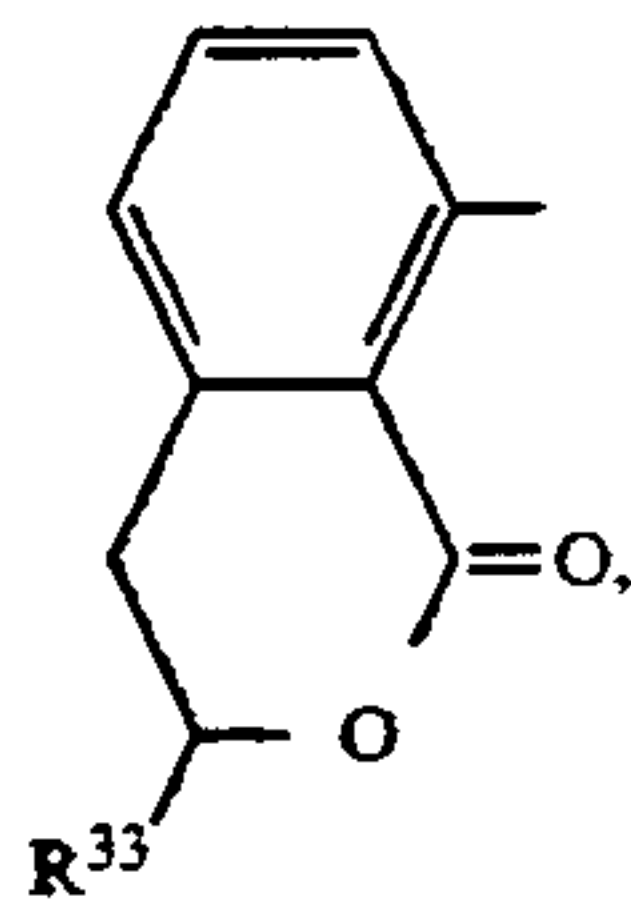


where

R³¹ represents C₁-C₄-alkyl and

R³² represents C₁-C₄-alkyl; where furthermore

R¹ represents the radical



where

R³³ represents hydrogen or methyl in which further-
more

R² represents hydrogen or the group —SO₂—R¹,
where

R¹ has the meaning indicated above as preferred;
in which furthermore

R³ represents hydrogen, fluorine, chlorine, bromine,
iodine, hydroxyl, mercapto, amino or an optionally
fluorine- and/or chlorine-substituted radical from the
series comprising C₁–C₄-alkyl, C₃–C₆-cycloalkyl, ben-
zyl, phenyl, C₁–C₄-alkoxy, C₃–C₄-alkenyloxy, C₃–C₄-
alkinyloxy, C₁–C₄-alkylthio, C₁–C₄-alkylsulphinyl,
C₁–C₄-alkylsulphonyl, C³–C⁴-alkenylthio, C³–C⁴-alki-
nylthio, benzyloxy, benzylthio, C₁–C₄-alkylamino and
di-(C₁–C₄-alkyl)-amino,

R⁴ represents hydrogen, fluorine, chlorine, bromine,
hydroxyl, amino, C₁–C₄-alkyl, C₁–C₄-halogenoalkyl,
C₁–C₂-alkoxy-C₁–C₂-alkyl, C₁–C₄-alkoxy, C₁–C₄-
halogenoalkoxy, C₁–C₂-alkoxy-C₁–C₂-alkoxy, C₁–C₄-
alkylthio, C₁–C₄-halogenoalkylthio, C₁–C₄-alkylsulphi-
nyl, C₁–C₄-alkylsulphonyl, C₁–C₄-alkylamino or di-
(C₁–C₂-alkyl)-amino,

X represents nitrogen or a CH group,

Y represents nitrogen or a CR⁵ group where

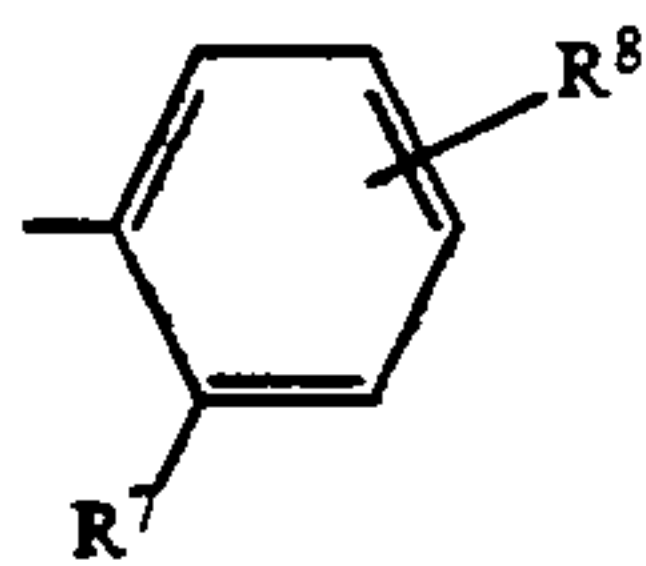
R⁵ represents hydrogen, fluorine, chlorine, bromine,
cyano, methyl, formyl, acetyl, methoxycarbonyl or
ethoxycarbonyl and

Z represents nitrogen or a CR⁶ group where

R⁶ represents hydrogen, fluorine, chlorine, bromine,
C₁–C₄-alkyl, C₁–C₄-alkoxy, C₁–C₄-halogenoalkoxy,
C₁–C₄-alkylthio, C₁–C₄-alkylsulphinyl, C₁–C₄-alkylsul-
phonyl, C₁–C₄-alkylamino, dimethylamino or diethyl-
amino.

The invention particularly relates to compounds of
the formula (I) in which

R¹ represents the radical

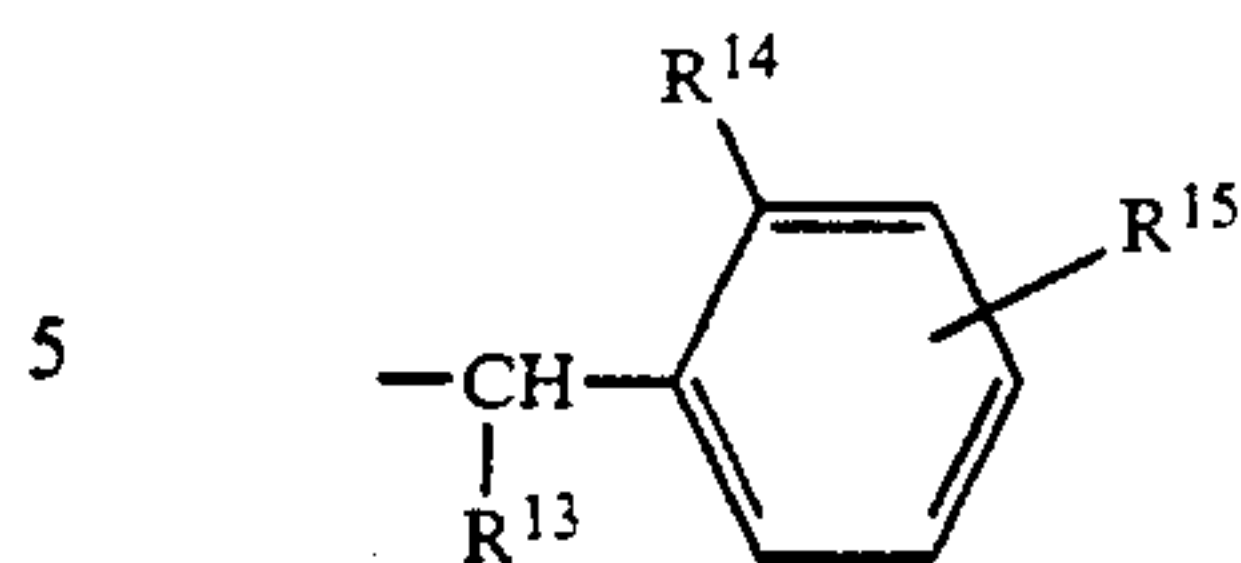


where

R⁷ represents fluorine, chlorine, bromine, methyl,
trifluoromethyl, methoxy, difluoromethoxy, tri-
fluoromethoxy, C₁–C₃-alkylthio, C₁–C₃-alkylsulphinyl,
C₁–C₃-alkylsulphonyl, dimethylaminosulphonyl, N-
methoxy-N-methylaminosulphonyl, phenyl, phenoxy or
C₁–C₃-alkoxycarbonyl and

R⁸ represents hydrogen, fluorine or chlorine; where
furthermore

R¹ represents the radical



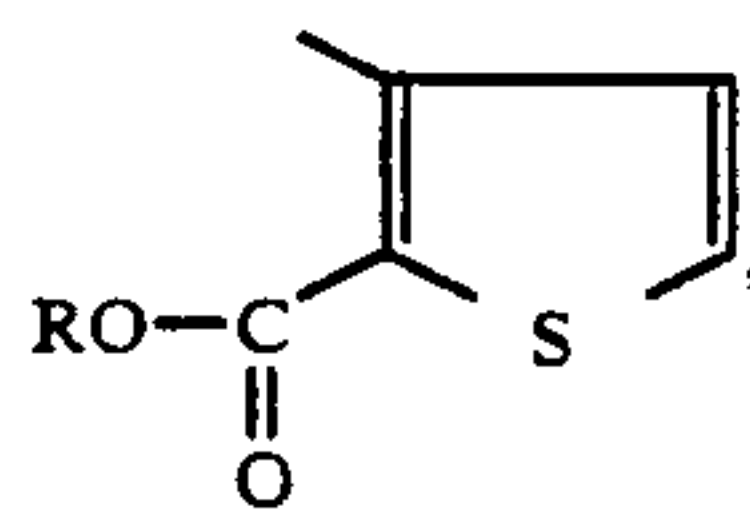
where

R¹³ represents hydrogen,

R¹⁴ represents fluorine, chlorine, bromine, methyl,
methoxy, difluoromethoxy, trifluoromethoxy, ethoxy,
methoxycarbonyl, ethoxycarbonyl, methylsulphonyl or
dimethylaminosulphonyl and

R¹⁵ represents hydrogen; where furthermore

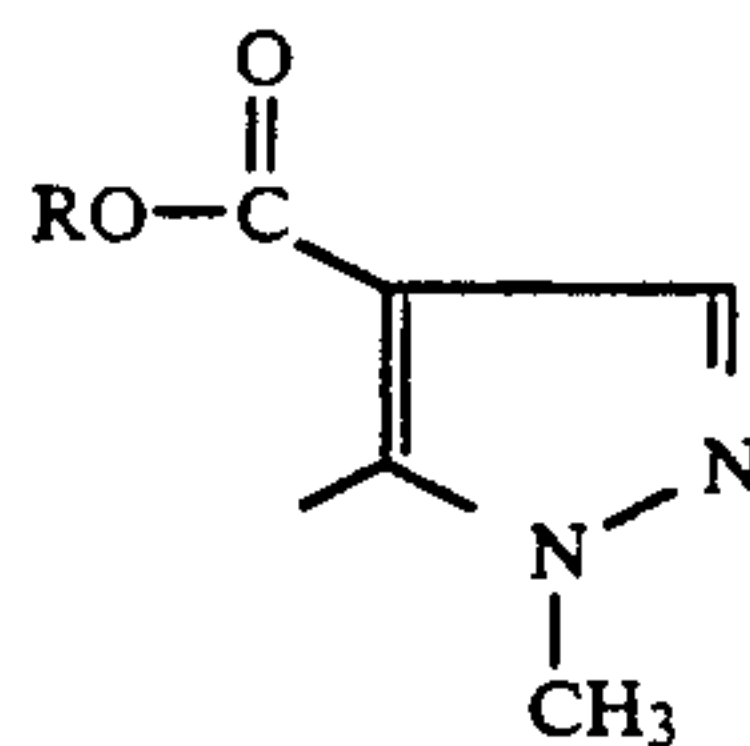
R¹ represents the radical



where

R represents C₁–C₄-alkyl, or

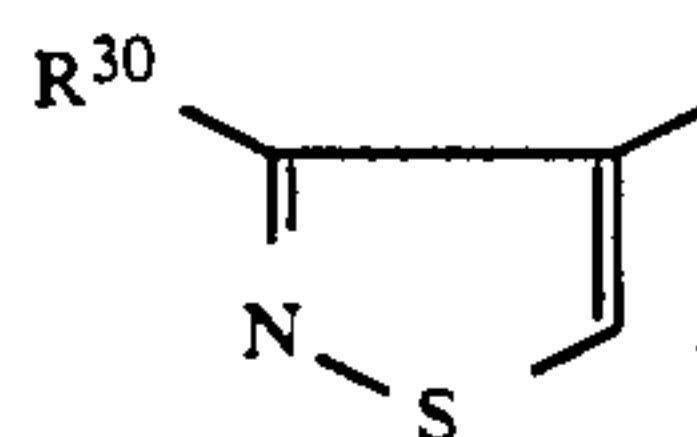
R¹ represents the radical



where

R represents C₁–C₄-alkyl, or

R¹ represents the radical



where

R³⁰ represents hydrogen, chlorine, methyl, ethyl,
methoxycarbonyl or ethoxycarbonyl;

in which furthermore

R² represents hydrogen or for the group —SO₂—R¹,

where

R¹ has the meaning indicated above as being particu-
larly preferred,

R³ represents hydrogen, methyl, ethyl, propyl, iso-
propyl, trifluoromethyl, cyclopropyl, benzyl, phenyl,
t-butyl, s-butyl, i-butyl, n-butyl, methoxy or methylthio,

R⁴ represents hydrogen, fluorine, chlorine, bromine,
methyl, trifluoromethyl, methoxy, ethoxy, difluorome-
thoxy, methylthio, ethylthio, amino, methylamino,
ethylamino, dimethylamino or diethylamino,

X represents nitrogen or a CH group,

Y represents nitrogen or a CR⁵ group where

R⁵ represents hydrogen, fluorine, chlorine or methyl,

and

Z represents nitrogen or a CR⁶ group where

R⁶ represents hydrogen, fluorine, chlorine, bromine,
methyl, ethyl, methoxy, ethoxy, propoxy, isopropoxy,

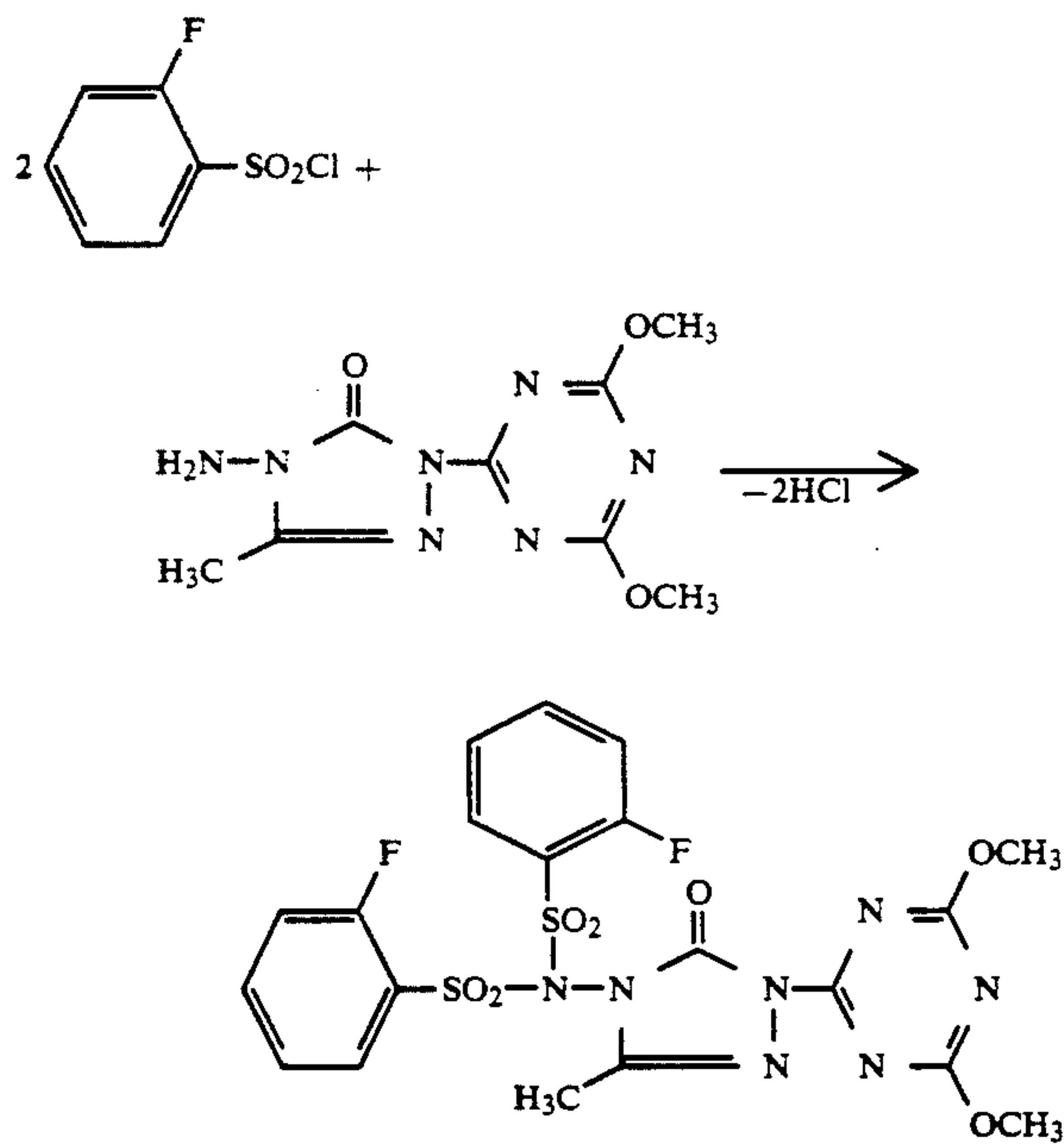
difluoromethoxy, methylthio, ethylthio, methylamino, ethylamino, dimethylamino or diethylamino.

The invention furthermore preferably relates to salts of compounds of the formula (I)

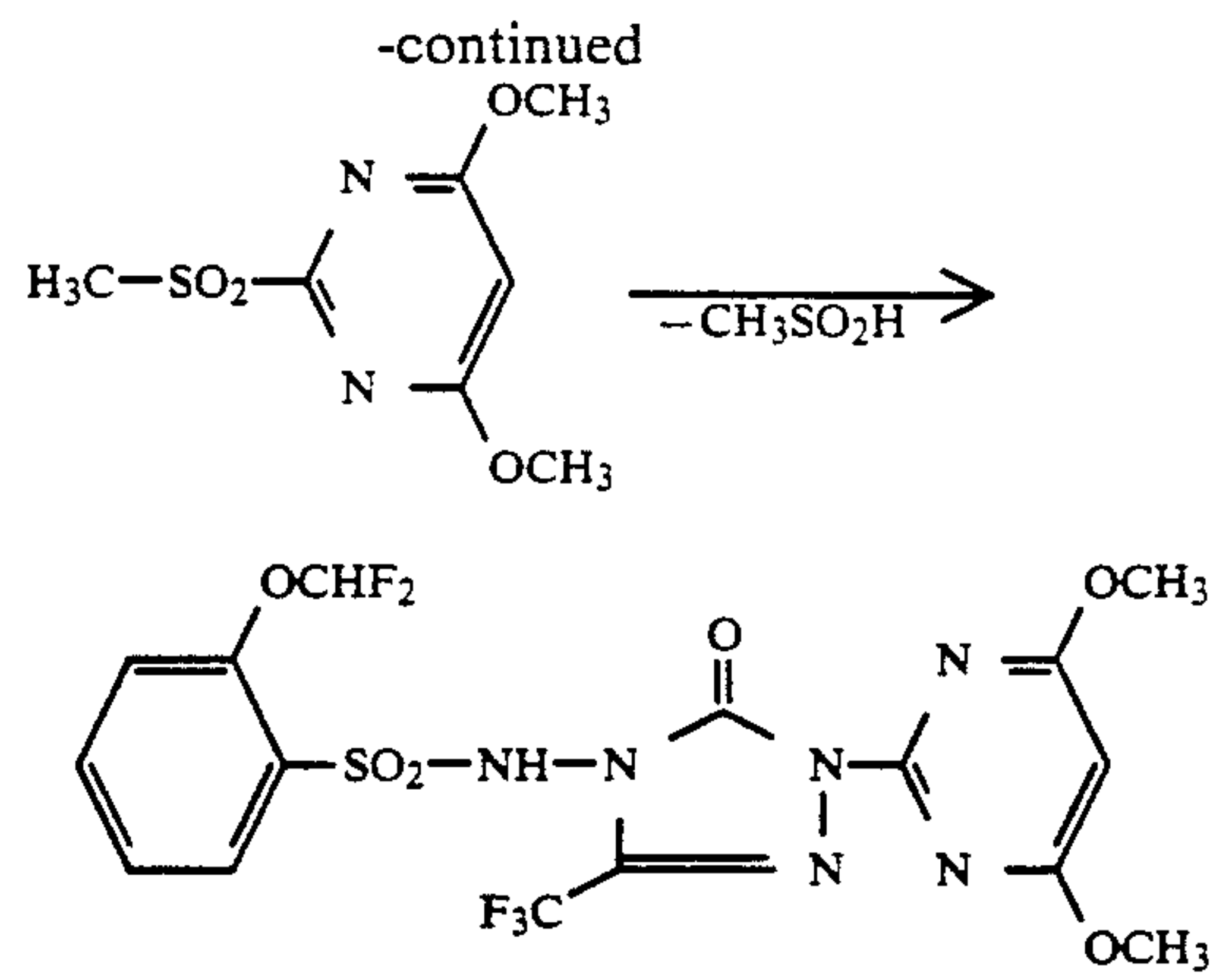
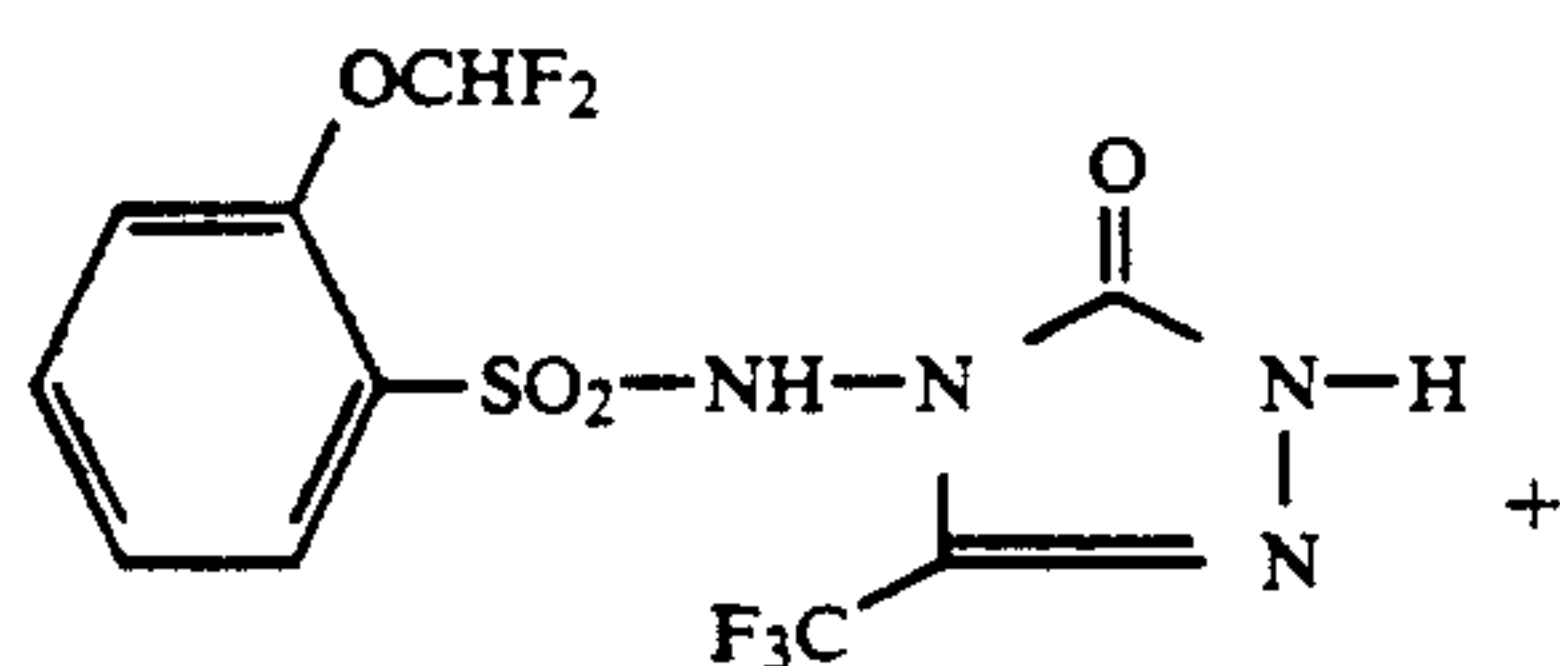
α) with proton acids, such as, for example, hydrochloric acid, sulphuric acid, phosphoric acid, methanesulphonic acid, benzene- or p-toluenesulphonic acid, or naphthalene-mono- or -di-sulphonic acids, or

β) with bases, such as, for example, sodium hydroxide, potassium hydroxide, calcium hydroxide, sodium hydride, potassium hydride, calcium hydride, sodium amide, potassium amide, calcium amide, sodium carbonate, potassium carbonate or calcium carbonate, sodium C₁-C₄-alkanoxides or potassium C₁-C₄-alkanoxides, ammonia, C₁-C₄-alkylamines, di-(C₁-C₄-alkyl)amines or tri-(C₁-C₄-alkyl)-amines.

If, for example, 4-amino-5-methyl-2-(4,6-dimethoxy-1,2,4-triazin-2-yl)-2,4-dihydro-3H-1,2,4-triazol-3-one and 2-fluorobenzenesulphonyl chloride (at least two mole equivalents) are used as starting substances in process (a) according to the invention, the course of the reaction in process (a) according to the invention can be outlined by the following equation:



If, for example, 4-(2-difluoromethoxy-phenylsulphonylamino)-5-trifluoromethyl-2,4-dihydro-3H-1,2,4-triazol-3-one and 4,6-dimethoxy-2-methylsulphonylpyrimidine are used as starting substances in process (b) according to the invention, the course of the reaction in process (b) according to the invention can be outlined by the following equation:



Formula (II) provides a general definition of the substituted aminotriazolones to be used as starting substances in the process (a) according to the invention for the preparation of compounds of the formula (I).

In formula (II), R³, R⁴, X, Y and Z preferably, or in particular, have those meanings which have already been mentioned above in connection with the description of the compounds of the formula (I) according to the invention as being preferred, or particularly preferred, for R³, R⁴, X, Y and Z.

Examples of the starting substances of the formula (II) are listed in Table 1 below.

TABLE 1

Examples of the starting substances of the formula (II)

R ³	R ⁴	X	Y	Z
H	CH ₃	N	CH	C-CH ₃
H	CH ₃	N	CH	C-OCH ₃
H	CH ₃	N	CH	C-OC ₂ H ₅
H	OCH ₃	N	CH	C-OCH ₃
H	OCH ₃	N	CH	C-Cl
H	H	N	CH	C-CH ₃
H	CF ₃	N	CH	C-CH ₃
H	CF ₃	N	CH	C-OCH ₃
H	CF ₃	N	CH	C-CF ₃
H	OCH ₃	N	CH	C-OCHF ₂
H	CH ₃	N	CH	C-OCHF ₂
H	OCHF ₂	N	CH	C-OCHF ₂
H	CH ₃	N	N	C-OCH ₃
H	OCH ₃	N	N	C-OCH ₃
H	OCH ₃	N	N	C-Cl
H	C ₂ H ₅	N	CH	C-OCH ₃
H	C ₂ H ₅	N	N	C-OCH ₃
CH ₃	CH ₃	N	CH	C-CH ₃
CH ₃	CH ₃	N	CH	C-OCH ₃
CH ₃	OCH ₃	N	CH	C-OCH ₃
CH ₃	OCH ₃	N	CH	C-Cl
CH ₃	OCHF ₂	N	CH	C-OCHF ₂
CH ₃	H	N	CH	C-CH ₃
CH ₃	CF ₃	N	CH	C-CH ₃
CH ₃	CF ₃	N	CH	C-OCH ₃
CH ₃	CF ₃	N	CH	C-CF ₃
CH ₃	OCH ₃	N	CH	C-OCHF ₂
CH ₃	Cl	N	CH	C-OCHF ₂
CH ₃	CH ₃	N	CH	C-OCHF ₂
CH ₃	NH-CH ₃	N	CH	C-OCH ₃
CH ₂	CH ₃	N	N	C-CH ₃
CH ₃	CH ₃	N	N	C-OCH ₃
CH ₃	OCH ₃	N	N	C-OCH ₃
CH ₃	C ₂ H ₅	N	CH	C-OCH ₃
CH ₃	CH ₃	N	N	C-OC ₂ H ₅
CH ₃	C ₂ H ₅	N	N	C-OCH ₃
CH ₃	CH ₃	N	N	C-Cl
CH ₃	CH ₃	CH	N	C-CH ₃
CH ₃	OCH ₃	CH	N	C-OCH ₃
CH ₃	CH ₃	N	CH	C-SCH ₃
H	CH ₃	N	CH	C-N(CH ₃) ₂
H	OCH ₃	N	CH	C-SCH ₃
H	OCH ₃	N	N	C-NHC ₂ H ₅
H	OC ₂ H ₅	N	N	C-NHCH ₃

TABLE 1-continued

Examples of the starting substances of the formula (II)				
R ³	R ⁴	X	Y	Z
H	CH ₃	CH	CH	C-CH ₃
OCH ₃	CH ₃	N	CH	C-CH ₃
OCH ₃	CH ₃	N	CH	C-OCH ₃
OCH ₃	OCH ₃	N	CH	C-OCH ₃
OCH ₃	OCH ₃	N	CH	C-Cl
OCH ₃	H	N	CH	C-CH ₃
OCH ₃	CF ₃	N	CH	C-OCH ₃
OCHF ₂	OCH ₃	N	CH	C-OCHF ₂
OCH ₃	CH ₃	N	CH	C-OCHF ₂
OCH ₃	CH ₃	N	N	C-CH ₃
OCH ₃	CH ₃	N	N	C-OCH ₃
OCH ₃	OCH ₃	N	N	C-OCH ₃
OCH ₃	C ₂ H ₅	N	CH	C-OCH ₃
OCH ₃	C ₂ H ₅	N	N	C-OCH ₃
SCH ₃	CH ₃	N	CH	C-CH ₃
OCH ₃	CH ₃	N	N	C-OC ₂ H ₅
SCH ₃	CH ₃	N	CH	C-OCH ₃
SCH ₃	CH ₃	N	CH	C-OC ₂ H ₅
SCH ₃	OCH ₃	N	CH	C-OCH ₃
SCH ₃	OCH ₃	N	CH	C-Cl
SCH ₃	H	N	CH	C-CH ₃
SCH ₃	CF ₃	N	CH	C-CF ₃
SCH ₃	CF ₃	N	CH	C-OCH ₃
SCH ₃	CF ₃	N	CH	C-CH ₃
SCH ₃	OCH ₃	N	CH	C-OCHF ₂
SCH ₃	CH ₃	N	CH	C-OCHF ₂
SCH ₃	OCHF ₂	N	CH	C-OCHF ₂
SCH ₃	NHCH ₃	N	CH	C-OCH ₃
SCH ₃	CH ₃	N	N	C-CH ₃
SCH ₃	CH ₃	N	N	C-OCH ₃
SCH ₃	OCH ₃	N	N	C-OCH ₃
SCH ₃	OCH ₃	N	N	C-Cl
SCH ₃	C ₂ H ₅	N	CH	C-OCH ₃
SCH ₃	C ₂ H ₅	N	N	C-OCH ₃
SCH ₃	OCH ₃	N	N	C-NH-C ₂ H ₅
SCH ₃	OC ₂ H ₅	N	N	C-NH-CH ₃
C ₂ H ₅	CH ₃	N	CH	C-CH ₃
C ₂ H ₅	CH ₃	N	CH	C-OCH ₃
C ₂ H ₅	CH ₃	N	CH	C-OC ₂ H ₅
C ₂ H ₅	OCH ₃	N	CH	C-OCH ₃
C ₂ H ₅	OCH ₃	N	CH	C-Cl
C ₂ H ₅	H	N	CH	C-CH ₃
C ₂ H ₅	CF ₃	N	CH	C-CH ₃
C ₂ H ₅	CF ₃	N	CH	C-OCH ₃
C ₂ H ₅	CF ₃	N	CH	C-CF ₃
C ₂ H ₅	OCH ₃	N	CH	C-OCHF ₂
C ₂ H ₅	CH ₃	N	CH	C-OCHF ₂
C ₂ H ₅	OCHF ₂	N	CH	C-OCHF ₂
C ₂ H ₅	NHCH ₃	N	CH	C-OCH ₃
C ₂ H ₅	CH ₃	N	N	C-CH ₃
C ₂ H ₅	CH ₃	N	N	C-OCH ₃
C ₂ H ₅	OCH ₃	N	N	C-OCH ₃
C ₂ H ₅	C ₂ H ₅	N	N	C-OCH ₃
C ₂ H ₅	CH ₃	N	N	C-N(CH ₃) ₂
C ₂ H ₅	OCH ₃	N	N	C-NHC ₂ H ₅
C ₂ H ₅	OC ₂ H ₅	N	N	C-NHCH ₃
C ₃ H _{7-n}	CH ₃	N	CH	C-CH ₃
C ₃ H _{7-n}	CH ₃	N	CH	C-OCH ₃
C ₃ H _{7-n}	CH ₃	N	CH	C-OC ₂ H ₅
C ₃ H _{7-n}	OCH ₃	N	CH	C-OCH ₃
C ₃ H _{7-n}	OCH ₃	N	CH	C-Cl
C ₃ H _{7-n}	H	N	CH	C-CH ₃
C ₃ H _{7-n}	CF ₃	N	CH	C-CH ₃
C ₃ H _{7-n}	CF ₃	N	CH	C-OCH ₃
C ₃ H _{7-n}	CF ₃	N	CH	C-CF ₃
C ₃ H _{7-n}	OCH ₃	N	CH	C-OCHF ₂
C ₃ H _{7-n}	CH ₃	N	CH	C-OCHF ₂
C ₃ H _{7-n}	OCHF ₂	N	CH	C-OCHF ₂
C ₃ H _{7-n}	NHCH ₃	N	CH	C-OCH ₃
C ₃ H _{7-n}	CH ₃	N	N	C-CH ₃
C ₃ H _{7-n}	CH ₃	N	N	C-OCH ₃
C ₃ H _{7-n}	OCH ₃	N	N	C-OCH ₃
C ₃ H _{7-n}	C ₂ H ₅	N	N	C-OCH ₃
C ₃ H _{7-n}	CH ₃	N	N	C-N(CH ₃) ₂
C ₃ H _{7-n}	OCH ₃	N	N	C-NHC ₂ H ₅
C ₃ H _{7-n}	OC ₂ H ₅	N	N	C-NHCH ₃
C ₃ H _{7-i}	CH ₃	N	CH	C-CH ₃
C ₃ H _{7-i}	CH ₃	N	CH	C-OCH ₃
C ₃ H _{7-i}	CH ₃	N	CH	C-OC ₂ H ₅

TABLE 1-continued








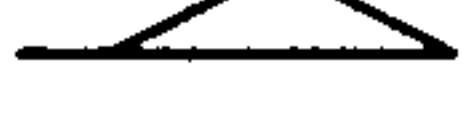







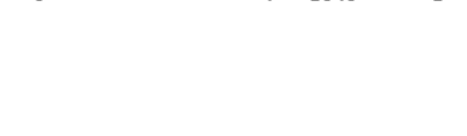




Examples of the starting substances of the formula (II)				
R ³	R ⁴	X	Y	Z
5 C ₃ H _{7-i}	OCH ₃	N	CH	C-OCH ₃
C ₃ H _{7-i}	OCH ₃	N	CH	C-Cl
C ₃ H _{7-i}	H	N	CH	C-CH ₃
C ₃ H _{7-i}	CF ₃	N	CH	C-CH ₃
C ₃ H _{7-i}	CF ₃	N	CH	C-OCH ₃
C ₃ H _{7-i}	CF ₃	N	CH	C-CF ₃
10 C ₃ H _{7-i}	OCH ₃	N	CH	C-OCHF ₂
C ₃ H _{7-i}	CH ₃	N	CH	C-OCHF ₂
C ₃ H _{7-i}	OCHF ₂	N	CH	C-OCHF ₂
C ₃ H _{7-i}	NHCH ₃	N	CH	C-OCH ₃
C ₃ H _{7-i}	CH ₃	N	N	C-CH ₃
C ₃ H _{7-i}	CH ₃	N	N	C-OCH ₃
15 C ₃ H _{7-i}	OCH ₃	N	N	C-OCH ₃
C ₃ H _{7-i}	C ₂ H ₅	N	N	C-OCH ₃
C ₃ H _{7-i}	CH ₃	N	N	C-N(CH ₃) ₂
C ₃ H _{7-i}	OCH ₃	N	N	C-NHC ₂ H ₅
C ₃ H _{7-i}	OC ₂ H ₅	N	N	C-NHCH ₃
20 	CH ₃	N	CH	C-CH ₃
	CH ₃	N	CH	C-OCH ₃
25 	CH ₃	N	CH	C-OC ₂ H ₅
	OCH ₃	N	CH	C-OCH ₃
30 	OCH ₃	N	CH	C-Cl
	H	N	CH	C-CH ₃
35 	CF ₃	N	CH	C-CH ₃
	CF ₃	N	CH	C-OCH ₃
40 	CF ₃	N	CH	C-CF ₃
	OCH ₃	N	CH	C-OCHF ₂
45 	CH ₃	N	CH	C-OCHF ₂
	OCHF ₂	N	CH	C-OCHF ₂
50 	NHCH ₃	N	CH	C-OCH ₃
	CH ₃	N	N	C-CH ₃
55 	CH ₃	N	N	C-OCH ₃
	OCH ₃	N	N	C-OCH ₃
60 	C ₂ H ₅	N	N	C-OCH ₃
	CH ₃	N	N	C-N(CH ₃) ₂
65 	OCH ₃	N	N	C-NHC ₂ H ₅
	OC ₂ H ₅	N	N	C-NHCH ₃
C ₄ H _{9-n}	CH ₃	N	CH	C-CH ₃

TABLE 1-continued

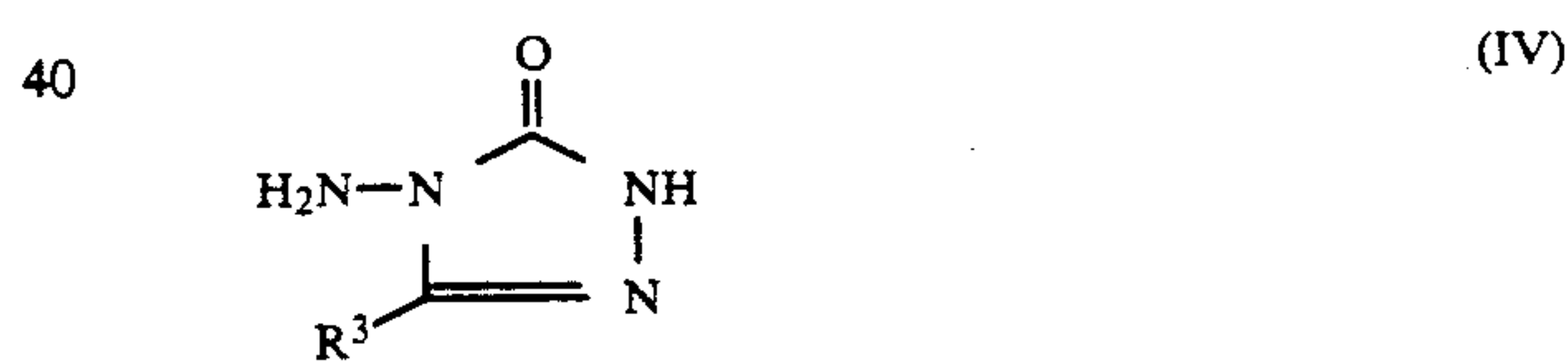
Examples of the starting substances of the formula (II)				
R ³	R ⁴	X	Y	Z
C ₄ H ₉ -n	CH ₃	N	CH	C-OCH ₃
C ₄ H ₉ -n	CH ₃	N	CH	C-OC ₂ H ₅
C ₄ H ₉ -n	OCH ₃	N	CH	C-OCH ₃
C ₄ H ₉ -n	OCH ₃	N	CH	C-Cl
C ₄ H ₉ -n	H	N	CH	C-CH ₃
C ₄ H ₉ -n	CF ₃	N	CH	C-CH ₃
C ₄ H ₉ -n	CF ₃	N	CH	C-OCH ₃
C ₄ H ₉ -n	CF ₃	N	CH	C-CF ₃
C ₄ H ₉ -n	OCH ₃	N	CH	C-OCHF ₂
C ₄ H ₉ -n	CH ₃	N	CH	C-OCHF ₂
C ₄ H ₉ -n	OCHF ₂	N	CH	C-OCHF ₂
C ₄ H ₉ -n	NHCH ₃	N	CH	C-OCH ₃
C ₄ H ₉ -n	CH ₃	N	N	C-CH ₃
C ₄ H ₉ -n	CH ₃	N	N	C-OCH ₃
C ₄ H ₉ -n	OCH ₃	N	N	C-OCH ₃
C ₄ H ₉ -n	C ₂ H ₅	N	N	C-OCH ₃
C ₄ H ₉ -n	CH ₃	N	N	C-N(CH ₃) ₂
C ₄ H ₉ -n	OCH ₃	N	N	C-NHC ₂ H ₅
C ₄ H ₉ -n	OC ₂ H ₅	N	N	C-NHCH ₃
C ₄ H ₉ -s	CH ₃	N	CH	C-CH ₃
C ₄ H ₉ -s	CH ₃	N	CH	C-OCH ₃
C ₄ H ₉ -s	CH ₃	N	CH	C-OC ₂ H ₅
C ₄ H ₉ -s	OCH ₃	N	CH	C-OCH ₃
C ₄ H ₉ -s	OCH ₃	N	CH	C-Cl
C ₄ H ₉ -s	H	N	CH	C-CH ₃
C ₄ H ₉ -s	CF ₃	N	CH	C-CH ₃
C ₄ H ₉ -s	CF ₃	N	CH	C-OCH ₃
C ₄ H ₉ -s	CF ₃	N	CH	C-CF ₃
C ₄ H ₉ -s	OCH ₃	N	CH	C-OCHF ₂
C ₄ H ₉ -s	CH ₃	N	CH	C-OCHF ₂
C ₄ H ₉ -s	OCHF ₂	N	CH	C-OCHF ₂
C ₄ H ₉ -s	NHCH ₃	N	CH	C-OCH ₃
C ₄ H ₉ -s	CH ₃	N	N	C-CH ₃
C ₄ H ₉ -s	CH ₃	N	N	C-OCH ₃
C ₄ H ₉ -s	OCH ₃	N	N	C-OCH ₃
C ₄ H ₉ -s	C ₂ H ₅	N	N	C-OCH ₃
C ₄ H ₉ -s	CH ₃	N	N	C-N(CH ₃) ₂
C ₄ H ₉ -s	OCH ₃	N	N	C-NHC ₂ H ₅
C ₄ H ₉ -s	OC ₂ H ₅	N	N	C-NHCH ₃
C ₄ H ₉ -i	CH ₃	N	CH	C-CH ₃
C ₄ H ₉ -i	CH ₃	N	CH	C-OCH ₃
C ₄ H ₉ -i	CH ₃	N	CH	C-OC ₂ H ₅
C ₄ H ₉ -i	OCH ₃	N	CH	C-OCH ₃
C ₄ H ₉ -i	OCH ₃	N	CH	C-Cl
C ₄ H ₉ -i	H	N	CH	C-CH ₃
C ₄ H ₉ -i	CF ₃	N	CH	C-CH ₃
C ₄ H ₉ -i	CF ₃	N	CH	C-OCH ₃
C ₄ H ₉ -i	CF ₃	N	CH	C-CF ₃
C ₄ H ₉ -i	OCH ₃	N	CH	C-OCHF ₂
C ₄ H ₉ -i	CH ₃	N	CH	C-OCHF ₂
C ₄ H ₉ -i	OCHF ₂	N	CH	C-OCHF ₂
C ₄ H ₉ -i	NHCH ₃	N	CH	C-OCH ₃
C ₄ H ₉ -i	CH ₃	N	N	C-CH ₃
C ₄ H ₉ -i	CH ₃	N	N	C-OCH ₃
C ₄ H ₉ -i	OCH ₃	N	N	C-OCH ₃
C ₄ H ₉ -i	C ₂ H ₅	N	N	C-OCH ₃
C ₄ H ₉ -i	CH ₃	N	N	C-N(CH ₃) ₂
C ₄ H ₉ -i	OCH ₃	N	N	C-NHC ₂ H ₅
C ₄ H ₉ -i	OC ₂ H ₅	N	N	C-NHCH ₃
C ₄ H ₉ -t	CH ₃	N	CH	C-CH ₃
C ₄ H ₉ -t	CH ₃	N	CH	C-OCH ₃
C ₄ H ₉ -t	CH ₃	N	CH	C-OC ₂ H ₅
C ₄ H ₉ -t	OCH ₃	N	CH	C-OCH ₃
C ₄ H ₉ -t	OCH ₃	N	CH	C-Cl
C ₄ H ₉ -t	H	N	CH	C-CH ₃
C ₄ H ₉ -t	CF ₃	N	CH	C-CH ₃
C ₄ H ₉ -t	CF ₃	N	CH	C-OCH ₃
C ₄ H ₉ -t	CF ₃	N	CH	C-CF ₃
C ₄ H ₉ -t	OCH ₃	N	CH	C-OCHF ₂
C ₄ H ₉ -t	CH ₃	N	CH	C-OCHF ₂
C ₄ H ₉ -t	OCHF ₂	N	CH	C-OCHF ₂
C ₄ H ₉ -t	NHCH ₃	N	CH	C-OCH ₃
C ₄ H ₉ -t	CH ₃	N	N	C-CH ₃
C ₄ H ₉ -t	CH ₃	N	N	C-OCH ₃
C ₄ H ₉ -t	OCH ₃	N	N	C-OCH ₃
C ₄ H ₉ -t	C ₂ H ₅	N	N	C-OCH ₃
C ₄ H ₉ -t	CH ₃	N	N	C-N(CH ₃) ₂
C ₄ H ₉ -t	OCH ₃	N	N	C-NHC ₂ H ₅
C ₄ H ₉ -t	OC ₂ H ₅	N	N	C-NHCH ₃

TABLE 1-continued

Examples of the starting substances of the formula (II)					
R ³	R ⁴	X	Y	Z	
5	CF ₃	CH ₃	N	CH	C-CH ₃
	CF ₃	CH ₃	N	CH	C-OCH ₃
	CF ₃	CH ₃	N	CH	C-OC ₂ H ₅
	CF ₃	OCH ₃	N	CH	C-OCH ₃
	CF ₃	OCH ₃	N	CH	C-Cl
	CF ₃	H	N	CH	C-CH ₃
	CF ₃	CF ₃	N	CH	C-CH ₃
10	CF ₃	CF ₃	N	CH	C-OCH ₃
	CF ₃	CF ₃	N	CH	C-CF ₃
	CF ₃	OCH ₃	N	CH	C-OCHF ₂
	CF ₃	CH ₃	N	CH	C-OCHF ₂
	CF ₃	OCHF ₂	N	CH	C-OCHF ₂
	CF ₃	NHCH ₃	N	CH	C-OCH ₃
15	CF ₃	CH ₃	N	N	C-CH ₃
	CF ₃	CH ₃	N	N	C-OCH ₃
	CF ₃	OCH ₃	N	N	C-OCH ₃
	CF ₃	OCH ₃	N	N	C-OCH ₃
	CF ₃	C ₂ H ₅	N	N	C-OCH ₃
	CF ₃	CH ₃	N	N	C-N(CH ₃) ₂
	CF ₃	OCH ₃	N	N	C-NHC ₂ H ₅
20	CF ₃	OC ₂ H ₅	N	N	C-NHCH ₃
	C ₆ H ₅	CH ₃	N	CH	C-CH ₃
	C ₆ H ₅	CH ₃	N	CH	C-OCH ₃
	C ₆ H ₅	OCH ₃	N	CH	C-OCH ₃
	C ₆ H ₅	OCH ₃	N	CH	C-Cl
25	C ₆ H ₅	CH ₃	N	N	C-OCH ₃
	C ₆ H ₅	OCH ₃	N	N	C-OCH ₃
	CH ₂ C ₆ H ₅	CH ₃	N	CH	C-CH ₃
	CH ₂ C ₆ H ₅	CH ₃	N	CH	C-OCH ₃
	CH ₂ C ₆ H ₅	OCH ₃	N	CH	C-OCH ₃
	CH ₂ C ₆ H ₅	OCH ₃	N	CH	C-Cl
	CH ₂ C ₆ H ₅	CH ₃	N	N	C-OCH ₃
30	CH ₂ C ₆ H ₅	OCH ₃	N	N	C-OCH ₃

The substituted aminotriazolinones of the formula (II), which are to be used as starting substances in process (a), were hitherto unknown from the literature. The new compounds of the formula (II) are obtained when

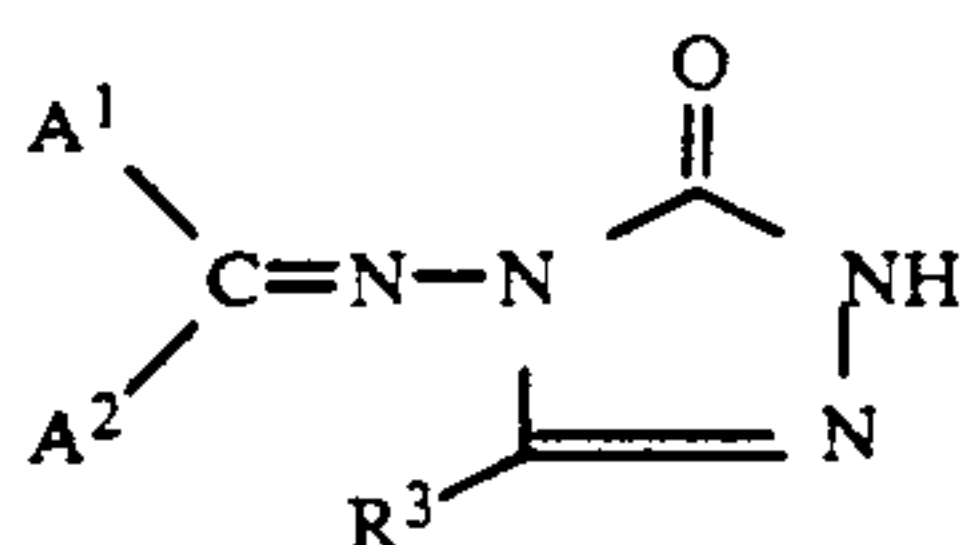
(a) aminotriazolinones of the general formula (VI)



45 in which R³ has the abovementioned meaning are reacted at temperatures between 20° C. and 150° C., if appropriate, with carbonyl compounds of the general formula (VII)

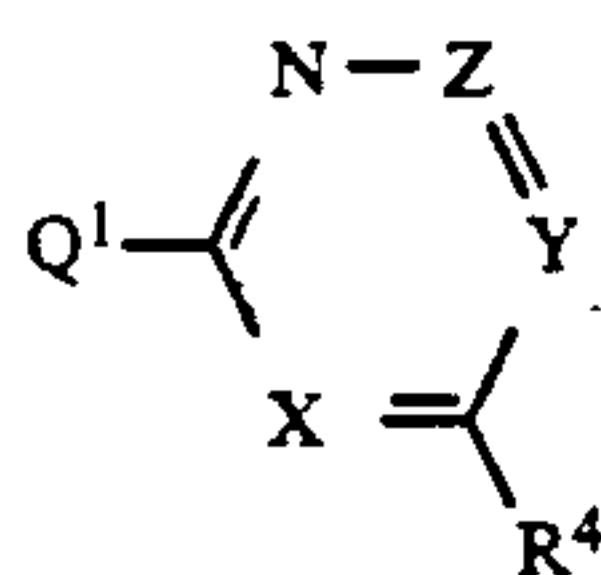


in which A¹ represents hydrogen or alkyl and A² represents alkyl or aryl, if appropriate in the presence of a catalyst, such as, for example, sulphuric acid, methanesulphonic acid or p-toluenesulphonic acid, and if appropriate in the presence of a diluent, such as, for example, methylene chloride, toluene, xylene, methanol, ethanol or isopropanol, the alkylideneaminotriazolinones (some of which are new) formed in this process of the general formula (VIII)



in which

A¹, A² and R³ have the abovementioned meanings, are, if appropriate, isolated and/or reacted virtually in situ, that is to say after concentrating the reaction mixture, with azines of the general formula (V)



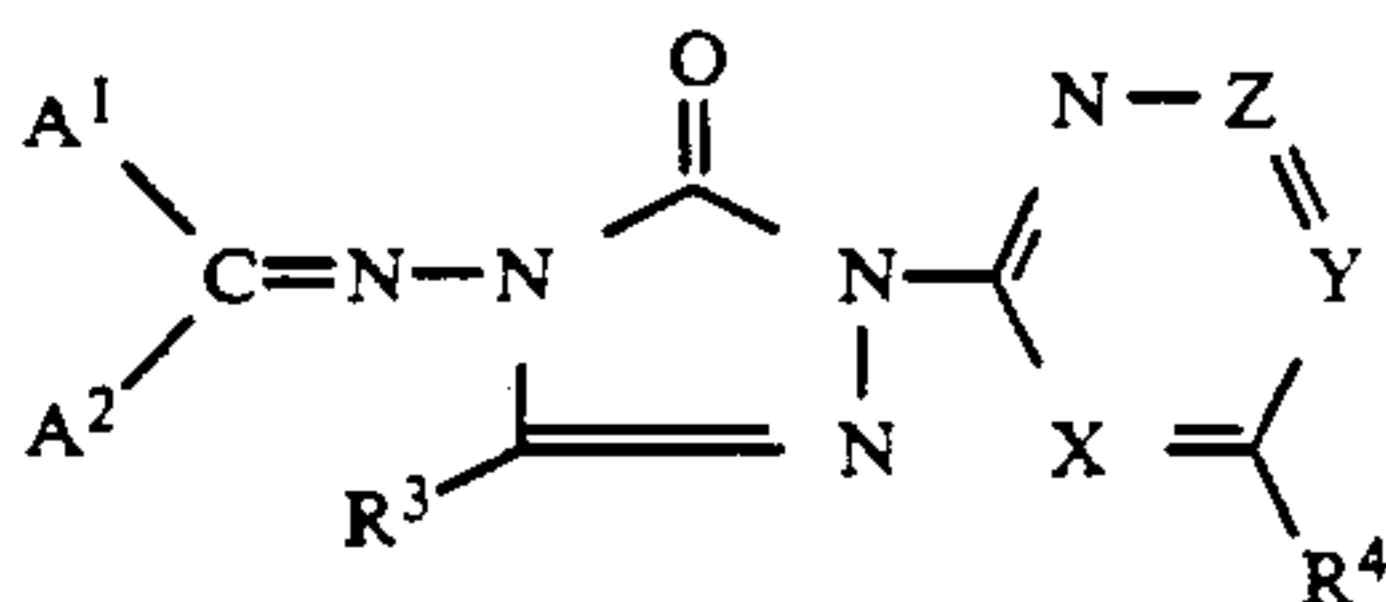
in which

R⁴, X, Y and Z have the abovementioned meanings and

Q¹ represents halogen, benzylsulphonyl or alkylsulphonyl,

if appropriate in the presence of an acid acceptor, such as, for example, potassium carbonate, and if appropriate in the presence of a diluent, such as, for example, acetonitrile or dimethylformamide, at temperatures between 0° C. and 150° C.

and the (new) substituted alkylideneaminotriazolones formed in this process, of the general formula (IX)

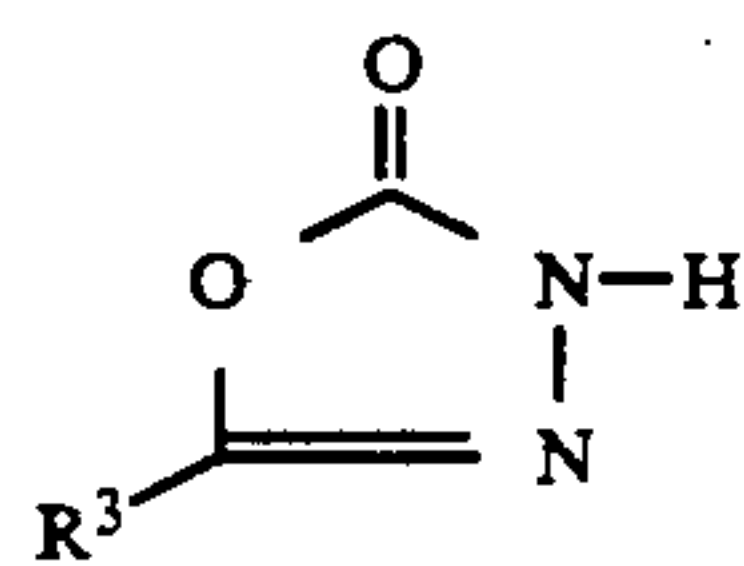


in which

A¹, A², A³, R⁴, X, Y and Z have the abovementioned meanings,

are hydrolyzed in a customary manner, for example by reacting them with water, if appropriate in the presence of an organic solvent, such as, for example, methanol, ethanol, isopropanol or dioxane, and if appropriate in the presence of an acid, such as, for example, hydrochloric acid or sulphuric acid, at temperatures between 20° C. and 100° C. to give the compounds of the formula (II); or when

(β) oxa-diazolones of the general formula (X)



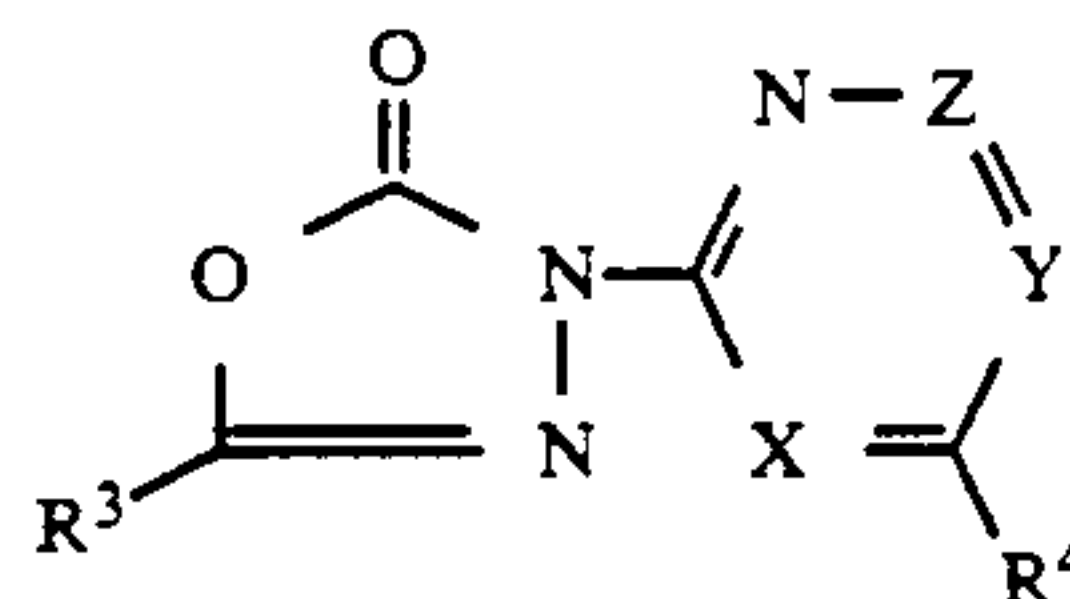
in which

R³ has the abovementioned meaning, are reacted with azines of the general formula (V) above

if appropriate in the presence of an acid acceptor, such as, for example, potassium carbonate, and if appropriate in the presence of a diluent, such as, for example,

acetonitrile, dioxane or dimethylformamide, at temperatures between 0° C. and 150° C.

and the (new) substituted oxadiazolones formed in this process, of the general formula (XI)



in which

R³, R⁴, X, Y and Z have the abovementioned meanings,

are reacted with hydrazine or hydrazine hydrate at temperatures between 0° C. and 150° C.

Some of the compounds of the formula (II) can be prepared using a synthesis route similar to the one described under (α) but without introducing protective groups using carbonyl compounds of the formula (VII), that is to say by directly reacting compounds of the formula (VI) with compounds of the formula (V) under the reaction condition indicated under (α).

Formula (VI) provides a general definition of the aminotriazolones, which are required as intermediates. In formula (VI), R³ preferably, or in particular, has the meaning which has already been indicated above in connection with the description of the compounds of the formula (I) according to the invention as being preferred, or particularly preferred, for R³.

Example of the compounds of the formula (VI) which may be mentioned are:

4-amino-2,4-dihydro-3H-1,2,4-triazol-3-one, 4-amino-5-methyl-2,4-dihydro-3H-1,2,4-triazol-3-one, 4-amino-5-methoxy-2,4-dihydro-3H-1,2,4-triazol-3-one, 4-amino-5-methylthio-2,4-dihydro-3H-1,2,4-triazol-3-one, 4-amino-5-ethyl-2,4-dihydro-3H-1,2,4-triazol-3-one, 4-amino-5-propyl-2,4-dihydro-3H-1,2,4-triazol-3-one, 4-amino-5-cyclopropyl-2,4-dihydro-3H-1,2,4-triazol-3-one, 4-amino-5-isopropyl-2,4-dihydro-3H-1,2,4-triazol-3-one, 4-amino-5-butyl-2,4-dihydro-3H-1,2,4-triazol-3-one, 4-amino-5-isobutyl-2,4-dihydro-3H-1,2,4-triazol-3-one and 4-amino-5-tert.-butyl-2,4-dihydro-3H-1,2,4-triazol-3-one.

The aminotriazolones of the formula (VI) are known and/or can be prepared by processes known per se (cf. Chem. Ber. 98 (1965), 3025-3033; J.Heterocycl. Chem. 21 (1984), 1769-1774; Doga: Kim. Ser. 10 (1986), 34-39—quoted in Chem. Abstracts 106: (1987), 138338e).

Formula (VII) provides a definition of the carbonyl compounds also required as intermediates.

In formula (VII),

A¹ preferably represents hydrogen or methyl and A² preferably represents methyl, ethyl, propyl, isopropyl, butyl, isobutyl or phenyl.

Examples of the compounds of the formula (VII) which may be mentioned are:

acetone, methyl ethyl ketone, methyl propyl ketone, methyl isopropyl ketone, methyl butyl ketone, methyl isobutyl ketone and benzaldehyde.

The intermediates of the formula (VII) are known chemicals for organic synthesis.

Formula (V) provides a general definition of the azines also required as intermediates. In formula (V), R⁴, X, Y and Z preferably, or in particular, have those meanings which have already been mentioned above in

connection with the description of the compounds of the formula (I) according to the invention as being preferred, or particularly preferred, for R⁴, X, Y and Z, and Q¹ preferably represents fluorine, chlorine, bromine, benzylsulphonyl or C₁-C₄-alkylsulphonyl, in particular represents chlorine or methylsulphonyl.

Examples of the compounds of the formula (V) which may be mentioned are:

2-chloro-, 2-benzylsulphonyl- and 2-methylsulphonyl-4,6-dimethyl-pyrimidine, -4-methyl-6-methoxy-pyrimidine, -4,6-dimethoxy-pyrimidine, -4-methyl-6-ethoxy-pyrimidine, -4-chloro-6-methoxy-pyrimidine, -4-methyl-pyrimidine, -4-chloro-6-methyl-pyrimidine, -4-trifluoromethyl-6-methoxy-pyrimidine, -4-methoxy-6-difluoromethoxy-pyrimidine, -4-methyl-6-difluoromethoxy-pyrimidine, -4,6-bis-difluoromethoxy-pyrimidine, -4-chloro-6-ethoxy-pyrimidine, -4-chloro-6-difluoromethoxy-pyrimidine, -4-methoxy-5-methyl-pyrimidine, -4-trifluoromethyl-6-difluoromethoxy-pyrimidine, -4,6-diethoxy-pyrimidine, -4,5-dichloro-6-methyl-pyrimidine, -4-methyl-5-chloro-6-methoxy-pyrimidine, -4,6-dichloropyrimidine, -4-ethyl-6-methoxy-pyrimidine, -5-chloro-4,6-dimethoxy-pyrimidine, -4-methoxy-6-methylamino-pyrimidine and -4,6-bis-trifluoromethyl-pyrimidine, furthermore 2-chloro-4,6-dimethyl-s-triazine, 2-chloro-4-methyl-6-methoxy-s-triazine, 2-chloro-4,6-dimethoxy-s-triazine, 2,4-dichloro-6-methoxy-s-triazine, 2-chloro-4-ethyl-6-methoxy-s-triazine, 2-chloro-4-methyl-6-ethoxy-s-triazine, 2-chloro-4-ethoxy-6-methylamino-s-triazine, 2-chloro-4-methoxy-6-methylamino-s-triazine, 2-chloro-4-methoxy-6-ethylamino-s-triazine and 2-chloro-4-ethoxy-6-ethylamino-s-triazine.

The azines of the formula (V) are known and/or can be prepared by processes known per se (cf., J. Chem. Soc. 1957, 1830, 1833; J. Org. Chem. 26 (1961), 792; U.S. Pat. No. 3,308,119 and U.S. Pat. No. 4,711,959).

Formula (X) provides a general definition of the oxadiazolones also to be used as intermediates. In formula (X), R³ preferably, or in particular, has the meaning which has already been mentioned above in connection with the description of the compounds of the formula (I) according to the invention as being preferred, or particularly preferred, for R³.

Examples of the compounds of the formula (X) which may be mentioned are:

1,3,4-oxadiazol-2(3H)-one, 5-methyl-1,3,4-oxadiazol-2(3H)-one, 5-ethyl-1,3,4-oxadiazol-2(3H)-one, 5-propyl-1,3,4-oxadiazol-2(3H)-one, 5-isopropyl-1,3,4-oxadiazol-2(3H)-one, 5-butyl-1,3,4-oxadiazol-2(3H)-one, 5-trifluoromethyl-1,3,4-oxadiazol-2(3H)-one, 5-tert.-butyl-1,3,4-oxadiazol-2(3H)-one, 5-cyclopropyl-1,3,4-oxadiazol-2(3H)-one, 5-methoxy-1,3,4-oxadiazol-2(3H)-one and 5-methylthio-1,3,4-oxadiazol-2(3H)-one.

The oxadiazolones of the formula (X) are known and/or can be prepared by processes known per se (cf. Helv. Chim. Acta 55 (1972), 1174-1178).

Formula (III) provides a general definition of the sulphonyl halides or sulphonic anhydrides also to be used as starting substances in process (a) according to the invention for the preparation of the new compounds of the formula (I). In formula (III), R¹ preferably, or in particular, has the meaning which has been indicated above in connection with the description of the compounds of the formula (I) according to the invention as being preferred, or particularly preferred, and Q preferably represents chlorine.

Starting substances of the formula (III) which may be mentioned by way of example are:

benzenesulphonyl chloride, 2-chloro-, 3-chloro-, 4-chloro-, 2,5-dichloro-, 2-fluoro-, 4-fluoro-, 2-bromo-, 4-bromo-, 2-cyano-, 2-nitro-, 4-nitro-, 2-methyl-, 4-methyl-, 2-chloromethyl-, 2-trifluoromethyl-, 2-methoxy-, 4-methoxy-, 2-(2-methoxy-ethoxy)-, 2-methylsulphonyl-, 2-isopropoxycarbonyl-, 2-chloro-6-methyl-, 2-bromo-6-methyl-, 2-methylthio-, 2-trifluoromethylthio-, 2-difluoromethylthio-, 2-cyclopropyloxycarbonyl-, 2-phenoxy-, 2-difluoromethoxy-, 2-trifluoromethoxy-, 2-(2-chloroethoxy)-, 2-methylthiomethyl-, 2-dimethylaminosulphonyl-, 2-phenyl-, 2-methoxycarbonyl-, 2-ethoxycarbonyl-, 2-dimethylaminocarbonyl- and 2-diethylaminocarbonyl-benzenesulphonyl chloride and (2-chloro-phenyl)-, (2-cyano-phenyl)-, (2-methoxycarbonyl-phenyl)-, (2-trifluoromethoxy-phenyl)- and (2-difluoromethoxy-phenyl)-methanesulphonyl chloride, furthermore 1-methyl-4-methoxycarbonyl-pyrazole-5-sulphonyl chloride, 1-methyl-4-ethoxycarbonyl-pyrazole-5-sulphonyl chloride, 1-methyl-4-bromo-pyrazole-5-sulphonyl chloride, 2-methoxycarbonyl-thiophene-3-sulphonyl chloride, 3-trifluoromethyl-pyridine-2-sulphonyl chloride, 3-dimethylaminocarbonyl-pyridine-2-sulphonyl chloride, 3-dimethylaminocarbonyl-6-methyl-pyridine-2-sulphonyl chloride, 3-dimethylaminocarbonyl-6-chloro-pyridine-2-sulphonyl chloride and 1-(iso)-quinolinyl-4-ethoxycarbonyl-pyrazole-5-sulphonyl chloride.

The sulphonyl halides or sulphonic anhydrides of the formula (III) are known and/or can be prepared by processes known per se (cf. J. Org. Chem. 33 (1968), 2104; J. Org. Chem. 25 (1960), 1824; DE-AS (German Published Specification) 2,308,262; EP-OS (European Published Specification) 23,140, 23,141, 23,422, 35,893, 48,143, 51,466, 64,322, 70,041, 44,808 and 44,809; U.S. Pat. No. 2,929,820, 4,282,242; 4,348,220 and 4,372,778 and Angew. Chem. 93 (1981), 151).

Process (a) according to the invention for the preparation of the new compounds of the formula (I) is preferably carried out using diluents. Suitable diluents for this process are virtually all inert organic solvents. These preferably include aliphatic and aromatic, optionally halogenated hydrocarbons, such as pentane, hexane, heptane, cyclohexane, petroleum ether, benzene, ligroin, benzene, toluene, xylene, methylene chloride, ethylene chloride, chloroform, carbon tetrachloride, chlorobenzene and o-dichlorobenzene, ethers, such as diethyl ether, dibutyl ether, glycol dimethyl ether, diglycol dimethyl ether, tetrahydrofuran and dioxane, ketones, such as acetone, methyl ethyl ketone, methyl isobutyl ketone and methyl isobutyl ketone, esters, such as methyl acetate and ethyl acetate, nitriles, such as, for example, acetonitrile and propionitrile, amides, such as, for example, dimethylformamide, dimethylacetamide and N-methyl-pyrrolidone, and dimethyl sulphoxide, tetramethylene sulphone and hexamethylphosphoric triamide and pyridine.

Acid acceptors which can be employed in process (a) according to the invention are all acid-binding agents which can customarily be used for reactions of this type. Alkali metal hydroxides, such as, for example, sodium hydroxide and potassium hydroxide, alkaline earth metal hydroxides, such as, for example, calcium hydroxide, alkali metal carbonates and alkali metal alkoxides, such as sodium carbonate and potassium carbonate, sodium tert.-butoxide and potassium tert.-butoxide.

ide, furthermore aliphatic, aromatic or heterocyclic amines, for example triethylamine, trimethylamine, dimethylaniline, dimethylbenzylamine, pyridine, picoline, 1,5-diazabicyclo-[4,3,0]-non-5-ene (DBN), 1,8-diazabicyclo-[5,4,0]-undec-7-ene (DBU) and 1,4-diazabicyclo-[2,2,2]-octane (DABCO), are preferably suitable.

In process (a) according to the invention, the reaction temperatures can be varied within a relatively wide range. In general, the reaction is carried out at temperatures between -50°C . and $+50^{\circ}\text{C}$., preferably at temperatures between -40°C . and $+40^{\circ}\text{C}$.

For carrying out process (a) according to the invention, between 1 and 5 moles, preferably between 1 and 4 moles, of sulphonyl halide or sulphonic anhydride of the formula (III) are generally employed per mole of aminotriazolinone of the formula (II). In the case where disulphonylated compounds of the formula (I, $\text{R}^2 = -\text{SO}_2-\text{R}^1$) are to be prepared in a one-pot reaction, at least 2 moles of sulphonyl halide or sulphonic anhydride (III) are to be employed per mole of aminotriazolinone (II).

The reactants can be combined in any desired sequence. In a preferred embodiment of process (a) according to the invention, the starting substances of the formula (II) and (III) are stirred at room temperature with a diluent, and the acid acceptor is slowly metered into this mixture, if appropriate after cooling. The reaction mixture is then stirred until the reaction is complete.

Working up can be carried out in a customary manner, for example by washing with water—if appropriate after concentrating and/or diluting the mixture with a virtually water-immiscible organic solvent, such as, for example, methylene chloride—drying and filtering the mixture, and carefully distilling off the solvent from the filtrate. The crude product, which remains in the residue, can be purified further in a customary manner, for example by column chromatography and/or by recrystallization.

The compounds which can be obtained as described above of the formula (I) in which R^2 represents the group $-\text{SO}_2-\text{R}^1$ can be reacted to give compounds of the formula (I) in

which R^2 represents hydrogen by reaction with desulphonylating agents, if appropriate in the presence of diluents.

In this connection, desulphonylating agents are taken to mean substances which can eliminate a sulphonyl group from N,N-bis-sulphonyl-amino compounds. Suitable desulphonylating agents are above all alkali metal hydroxides or alkaline earth metal hydroxides, such as sodium hydroxide, potassium hydroxide and calcium hydroxide, alkali metal alkoxides, such as sodium methoxide, potassium methoxide, sodium ethoxide and potassium ethoxide, furthermore ammonia, alkylamines, such as methylamine, ethylamine, propylamine and butylamine, and also dialkylamines, such as dimethylamine and diethylamine. It is preferred to employ ammonia as the desulphonylating agent.

The desulphonylation is preferably carried out in the presence of diluents. Preferred diluents, besides water, are polar organic solvents, such as methanol, ethanol, propanol, isopropanol, 2-methoxyethanol, 2-ethoxyethanol and dioxane.

The desulphonylation is generally carried out at temperatures between 0°C . and 150°C ., preferably at temperatures between 20°C . and 100°C .

In general, the reactants for desulphonylation are mixed at room temperature and stirred until the reaction is complete, if appropriate at increased temperature. For working up, the mixture is concentrated, if appropriate, diluted with water, and acidified using a strong acid, such as, for example, hydrochloric acid. The product, which is obtained in this process in the form of crystals (I, $\text{R}^2 = \text{H}$), can be isolated by filtering off with suction.

If desired, salts can be prepared from the compounds of the general formula (I) according to the invention. Such salts are obtained in a simple manner by customary salt formation methods, for example by dissolving or dispersing a compound of the formula (I) in a suitable solvent, such as, for example, water, methanol, ethanol or acetone, and adding a suitable acid or base. The salts can then be isolated by concentration or filtering off with suction, if appropriate after stirring for some time.

Formula (IV) provides a general definition of the sulphonylaminotriazolinones to be used as starting substances in process (b) according to the invention for the preparation of compounds of the formula (I).

In formula (IV), R^1 , R^2 and R^3 preferably, or in particular, have the meanings which have already been indicated above in connection with the description of the compounds of the formula (I) according to the invention as being preferred, or particularly preferred, for R^1 , R^2 and R^3 .

Starting substances of the formula (IV) which may be mentioned by way of example are:

4-(phenylsulphonylamino)-, 4-(2-chloro-phenylsulphonylamino)-, 4-(2-fluoro-phenylsulphonylamino)-, 4-(2-bromophenylsulphonylamino)-, 4-(2-methyl-phenylsulphonylamino)-, 4-(2-methoxy-phenylsulphonylamino)-, 4-(2-trifluoromethyl-phenylsulphonylamino)-, 4-(2-methylthio-phenylsulphonylamino)-, 4-(2-methylsulphonyl-phenylsulphonylamino)-, 4-(2-isopropoxycarbonyl-phenylsulphonylamino)-, 4-(2-chloro-6-methyl-phenylsulphonylamino)-, 4-(2-phenyl-phenylsulphonylamino)-, 4-(2-difluoromethoxy-phenylsulphonylamino)-, 4-(2-trifluoromethoxy-phenylsulphonylamino)-, 4-(2-dimethylaminosulphonylphenylsulphonylamino)-, 4-(2-methoxycarbonyl-phenylsulphonylamino)-, 4-(2-ethoxycarbonyl-phenylsulphonylamino)-, 4-(2-chlorophenylmethylsulphonylamino)-, 4-(2-methoxycarbonyl-phenylmethylsulphonylamino)-, 4-(2-ethoxycarbonylphenylmethylsulphonylamino)-, 4-(2-difluoromethoxy-phenylmethylsulphonylamino)-, 4-(2-trifluoromethoxy-phenylmethylsulphonylamino)-, 4-(1-methyl-4-methoxycarbonyl-pyrazol-5-ylsulphonylamino)-, 4-(1-methyl-4-bromo-pyrazol-5-ylsulphonylamino)-, 4-(2-methoxycarbonyl-thiophen-3-ylsulphonylamino)-, 4-(3-trifluoromethyl-pyridin-2-ylsulphonylamino)-, 4-(3-dimethylaminocarbonyl-pyridin-2-ylsulphonylamino)- and 4-(1-isoquinolinyl-4-ethoxycarbonyl-pyrazol-5-ylsulphonylamino)-, 2,4-dihydro-3H-1,2,4-triazol-3-one, -5-methyl-2,4-dihydro-3H-1,2,4-triazol-3-one, -5-ethyl-2,4-dihydro-3H-1,2,4-triazol-3-one, -5-propyl-2,4-dihydro-3H-1,2,4-triazol-3-one, -5-isopropyl-2,4-dihydro-3H-1,2,4-triazol-3-one, -5-trifluoromethyl-2,4-dihydro-3H-1,2,4-triazol-3-one, -5-methoxy-2,4-dihydro-3H-1,2,4-triazol-3-one, -5-methylthio-2,4-dihydro-3H-1,2,4-triazol-3-one, -5-t-butyl-2,4-dihydro-3H-1,2,4-triazol-3-one, -5-benzyl-2,4-dihydro-3H-1,2,4-triazol-3-one and -5-phenyl-2,4-dihydro-3H-1,2,4-triazol-3-one.

methylene chloride, aliphatic hydrocarbons, such as cyclohexane or paraffins, for example petroleum fractions, mineral and vegetable oils, alcohols, such as butanol or glycol as well as their ethers and esters, ketones, such as acetone, methyl ethyl ketone, methyl isobutyl ketone or cyclohexanone, strongly polar solvents, such as dimethylformamide and dimethyl sulphoxide, as well as water.

As solid carriers there are suitable: for example ammonium salts and ground natural minerals, such as kaolins, clays, talc, chalk, quartz, attapulgite, montmorillonite or diatomaceous earth, and ground synthetic minerals, such as highly disperse silica, alumina and silicates, as solid carriers for granules there are suitable for example crushed and fractionated natural rocks such as calcite, marble, pumice, sepiolite and dolomite, as well as synthetic granules of inorganic and organic meals, and granules of organic material such as sawdust, coconut shells, corn cobs and tobacco stalks; as emulsifying and/or foam-forming agents there are suitable: for example non-ionic and anionic emulsifiers, such as polyoxyethylene-fatty acid esters, polyoxyethylene-fatty alcohol ethers, for example alkylaryl polyglycol ethers, alkylsulphonates, alkyl sulphates, arylsulphonates as well as albumin hydrolysis products; as dispersing agents there are suitable: for example lignin-sulphite waste liquors and methylcellulose.

Adhesives such as carboxymethylcellulose and natural and synthetic polymers in the form of powders, granules or latices, such as gum arabic, polyvinyl alcohol and polyvinyl acetate, as well as natural phospholipids, such as cephalins and lecithins, and synthetic phospholipids, can be used in the formulations. Further additives can be mineral and vegetable oils.

It is possible to use colorants such as inorganic pigments, for example iron oxide, titanium oxide and Prussian Blue, and organic dyestuffs, such as alizarin dyestuffs, azo dyestuffs and metal phthalocyanine dyestuffs, and trace nutrients such as salts of iron, manganese, boron, copper, cobalt, molybdenum and zinc.

The formulations in general contain between 0.1 and 95 per cent by weight of active compound, preferably between 0.5 and 90%.

The active compounds according to the invention, as such or in the form of their formulations, can also be used, for combating weeds, as mixtures with known herbicides, finished formulations or tank mixes being possible.

Suitable herbicides for the mixtures are known herbicides, such as, for example, 1-amino-6-ethylthio-3-(2,2-dimethylpropyl)-1,3,5-triazine-2,4(1H,3H)-dione (AMETHYDIONE) or N-(2-benzothiazolyl)-N,N'-dimethylurea (METABENZTHIAZURON) for combating weeds in cereals; 4-amino-3-methyl-6-phenyl-1,2,4-triazin-5(4H)-one (METAMITRON) for combating weeds in sugar beets and 4-amino-6-(1,1-dimethylethyl)-3-methylthio-1,2,4-triazin-5(4H)-one (METRIBUZIN) for combating weeds in soy beans; 2,4-dichlorophenoxyacetic acid (2,4-D); 4-(2,4-dichlorophenoxy)-butyric acid (2,4-DB); 2,4-dichlorophenoxypropionic acid (2,4-DP); 3-isopropyl-2,1,3-benzothiadiazin-4-one 2,2-dioxide (BENTAZONE); methyl 5-(2,4-dichlorophenoxy)-2-nitrobenzoate (BIFENOX); 3,5-di-bromo-4-hydroxy-benzonitrile (BROMOXYNIL); 2-chloro-N-[[[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)-amino]-carbonyl]benzenesulphonamide (CHLORSULFURON); N,N-dimethyl-N'-(3-chloro-4-methylphenyl)-urea (CHLORTOLURON); 2-[4-(2,4-dichlorophenoxy)-

phenoxy]-propionic acid, its methyl ester or its ethyl ester (DICLOFOP); 2-[4-[(6-chloro-2-benzoxazolyl)-oxy]-phenoxy]-propionic acid, its methyl ester or its ethyl ester (FENOXAPROP); [(4-amino-3,5-dichloro-6-fluoro-2-pyridinyl)-oxy]-acetic acid or its 1-methylheptyl ester (FLUROXYPYR); methyl 2-[4,5-dihydro-4-methyl-4-(1-methylethyl)-5-oxo-1H-imidazol-2-yl]-4(5)-methylbenzoate (IMAZAMETHABENZ); 3,5-diiodo-4-hydroxybenzonitrile (IOXYNIL); N,N-dimethyl-N'-(4-isopropylphenyl)-urea (ISOPROTURON); (2-methyl-4-chlorophenoxy)-acetic acid (MCPA); (4-chloro-2-methylphenoxy)-propionic acid (MCPPI); N-methyl-2-(1,3-benzothiazol-2-yloxy)-acetanilide (MEFENACET); 2-[[[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)-amino]-carbonyl]-amino]-sulphonyl]-benzoic acid or its methyl ester (METSULFURON); N-(1-ethylpropyl)-3,4-dimethyl-2,6-dinitroaniline (PENDINGIMETHALIN); 0-(6-chloro-3-phenyl-pyridazin-4-yl) S-octyl thiocarbonate (PYRIDATE); 2-[1-(ethoxamino)-butylidene]-5-(2-ethylthiopropyl)-1,3-cyclohexadione (SETHOXYDIM); 4-ethylamino-2-t-butylamino-6-methylthio-s-triazine (TERBUTRYNE) and methyl 3-[[[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)-amino]-carbonyl]amino]-sulphonyl]-thiophene-2-carboxylate (THIAMETURON). Surprisingly, some mixtures also show synergistic action.

Mixtures with other known active compounds, such as fungicides, insecticides, acaricides, nematocides, bird repellents, plant nutrients and agents which improve soil structure, are also possible.

The active compounds can be used as such, in the form of their formulations or in the use forms prepared therefrom by further dilution, such as ready-to-use solutions, suspensions, emulsions, powders, pastes and granules. They are used in the customary manner, for example by watering, spraying, atomizing or scattering.

The active compounds according to the invention can be applied either before or after emergence of the plants.

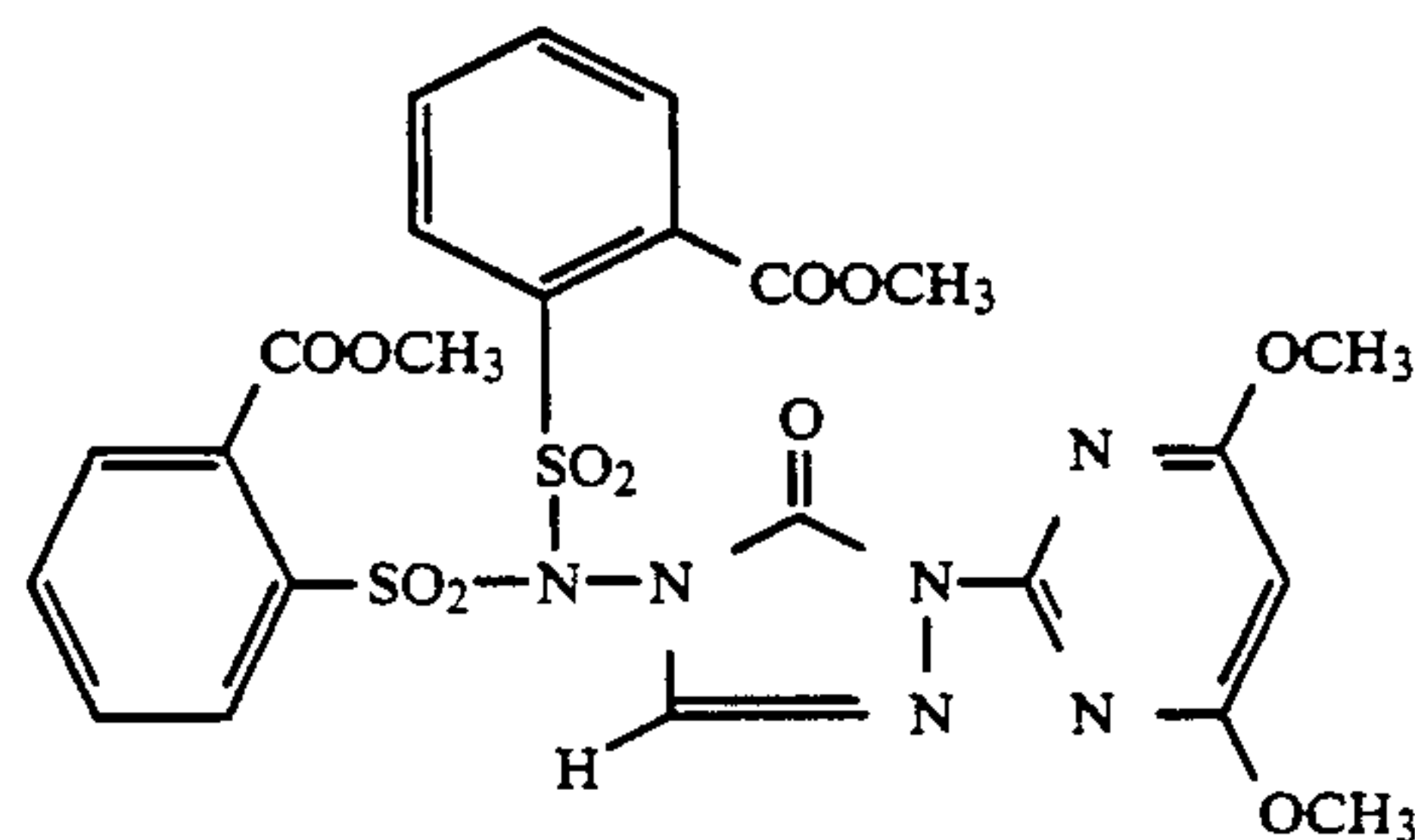
They can also be incorporated into the soil before sowing.

The amount of active compound used can vary within a substantial range. It depends essentially on the nature of the desired effect. In general, the amounts used are between 0.01 and 10 kg of active compound per hectare of soil surface, preferably between 0.05 and 5 kg per ha.

The preparation and use of the active compounds according to the invention can be seen from the following examples.

PREPARATION EXAMPLES

Example 1

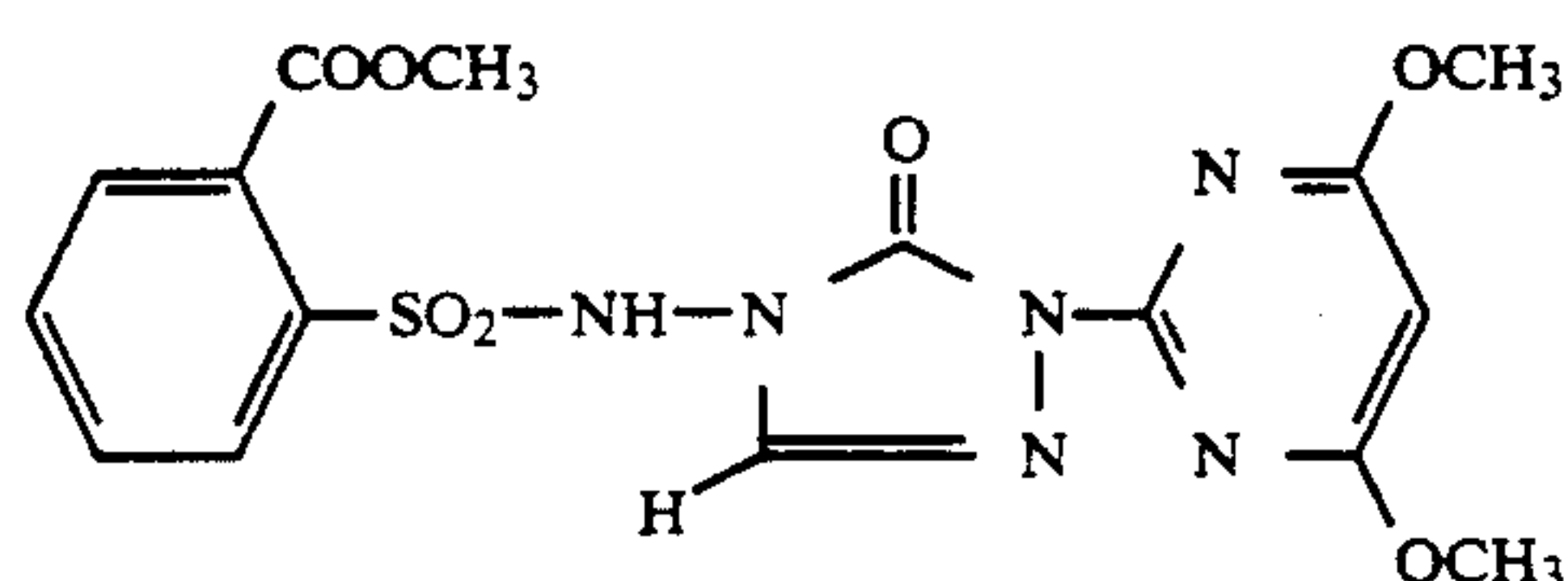


(Process (a))

A mixture of 7.2 g (0.03 mol) of 4-amino-2-(4,6-dimethoxypyrimidin-2-yl)-2,4-dihydro-3H-1,2,4-triazol-3-one, 21.0 g (0.09 mol) of 2-methoxycarbonylbenzenesulphonyl chloride and 50 ml of pyridine is stirred at 20° C. for 12 hours. The mixture is subsequently concentrated under a waterpump vacuum, and the residue is taken up in methylene chloride and washed with 2N hydrochloric acid. The mixture is dried using magnesium sulphate and then filtered, and the solvent is carefully distilled off from the filtrate under a waterpump vacuum.

12.0 g (63% of theory) of 4-[bis-(2-methoxycarbonylphenylsulphonyl)-amino]-2-(4,6-dimethoxy-pyrimidin-2-yl)-2,4-dihydro-3H-1,2,4-triazol-3-one are obtained as a crystalline residue of melting point 176° C.

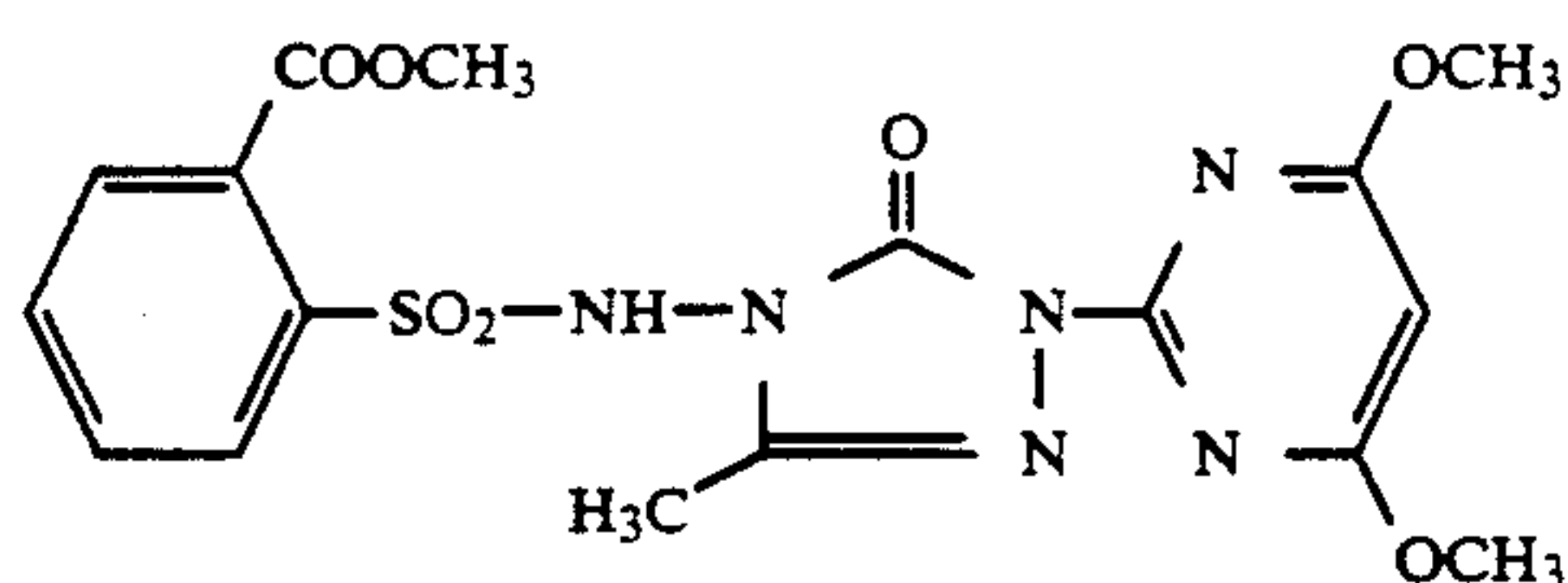
Example 2



A mixture of 6.4 g (0.01 mol) of 4-[bis-(2-methoxycarbonylphenylsulphonyl)-amino]-2-(4,6-dimethoxy-pyrimidin-2-yl)-2,4-dihydro-3H-1,2,4-triazol-3-one (cf. Example 1), 100 ml of methanol and 10 ml of concentrated aqueous ammonia is stirred at 50° C. for 60 minutes. The mixture is subsequently concentrated under a waterpump vacuum, and the residue is taken up in water and acidified using 2N hydrochloric acid. The product which is obtained in this process in the form of crystals is isolated by filtering off with suction.

4.0 g (92% of theory) of 4-(2-methoxycarbonylphenylsulphonylamino)-2-(4,6-dimethoxy-pyrimidin-2-yl)-2,4-dihydro-3H-1,2,4-triazol-3-one of melting point 192° C. are obtained.

Example 3



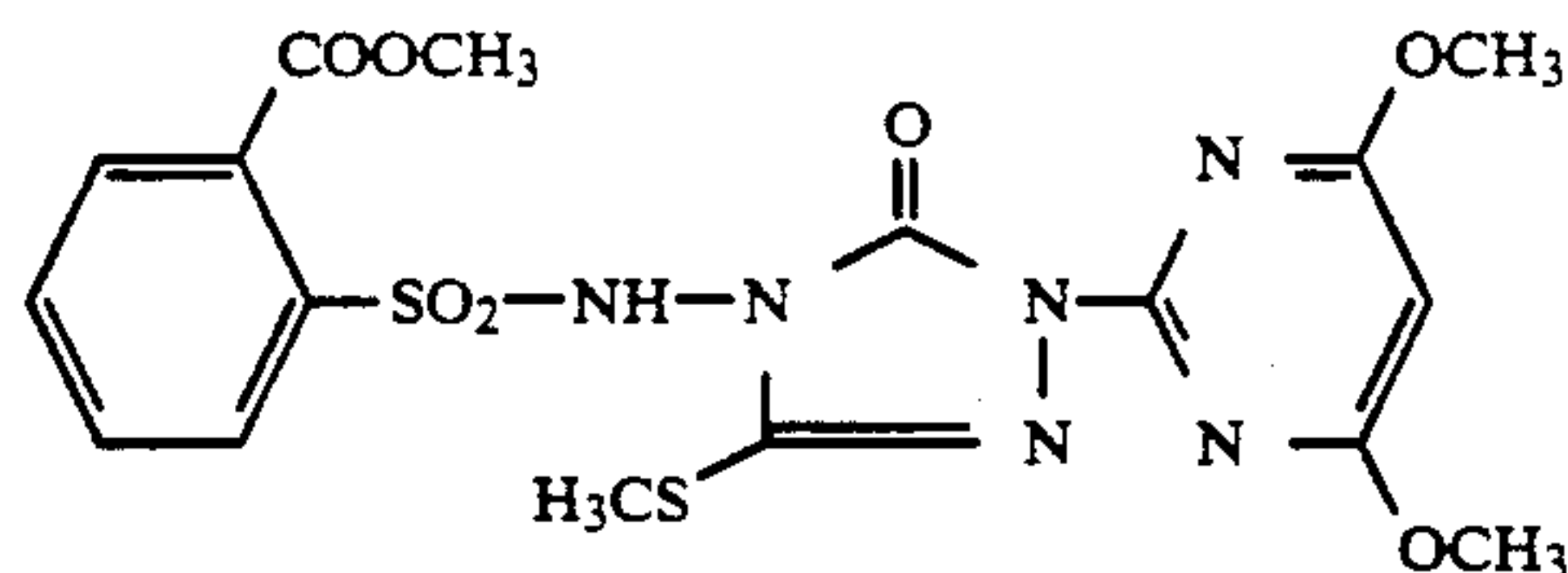
(Process (a))

A mixture of 4.2 g (0.016 mol) of 4-amino-5-methyl-2-(4,6-dimethoxy-pyrimidin-2-yl)-2,4-dihydro-3H-1,2,4-

triazol-3-one, 11.7 g (0.05 mol) of 2-methoxycarbonylbenzenesulphonyl chloride and 50 ml of pyridine is stirred at 20° C. for 12 hours, then diluted with ice-water and acidified using 2N hydrochloric acid, and the mixture is shaken with methylene chloride. The organic phase is separated off, dried using magnesium sulphate and filtered. The solvent is carefully distilled off from the filtrate under a waterpump vacuum.

3.8 g (50% of theory) of 4-(2-methoxycarbonylphenylsulphonylamino)-5-methyl-2-(4,6-dimethoxy-pyrimidin-2-yl)-2,4-dihydro-3H-1,2,4-triazol-3-one are obtained as a crystalline residue of melting point 174° C.

Example 4



(Process (a))

A mixture of 5.7 g (0.02 mol) of 4-amino-5-methylthio-2-(4,6-dimethoxy-pyrimidin-2-yl)-2,4-dihydro-3H-1,2,4-triazol-3-one, 14.0 g (0.06 mol) of 2-methoxycarbonylbenzenesulphonyl chloride and 50 ml of pyridine is stirred at 20° C. for 48 hours. The mixture is subsequently concentrated under a waterpump vacuum, the residue is taken up in methylene chloride, and the mixture is washed using 2N hydrochloric acid, dried using magnesium sulphate and filtered. The filtrate is concentrated and the residue is purified by column chromatography on silica gel using methylene chloride/methanol (vol. 10:1).

3.0 g (31% of theory) of 4-(2-methoxycarbonylphenylsulphonylamino)-5-methylthio-2-(4,6-dimethoxy-pyrimidin-2-yl)-2,4-dihydro-3H-1,2,4-triazol-3-one of melting point 179° C. are obtained.

The compounds of the formula (I) which are listed in Table 2 below can be prepared analogously to Examples 1 to 4 and in accordance with the general description of the process according to the invention.

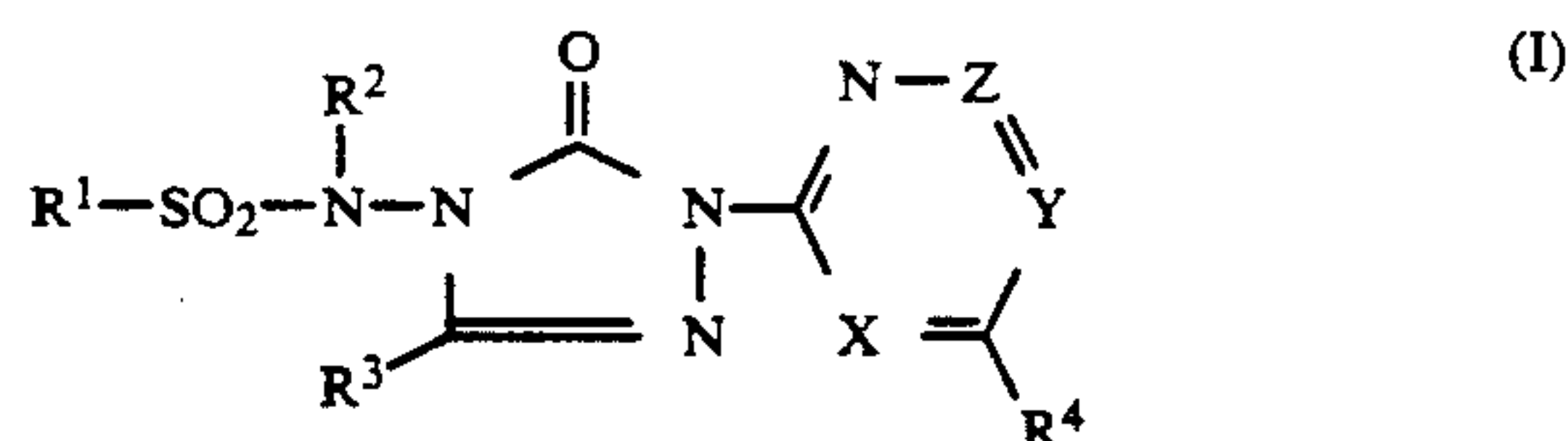


TABLE 2

Ex- am- ple No.	Examples of the compounds of the formula (I)								Melting point (°C.)
	R ¹	R ²	R ³	R ⁴	X	Y	Z		
5		H	H	SO ₂ CH ₃	N	CH	C—OCH ₃	191	

TABLE 2-continued

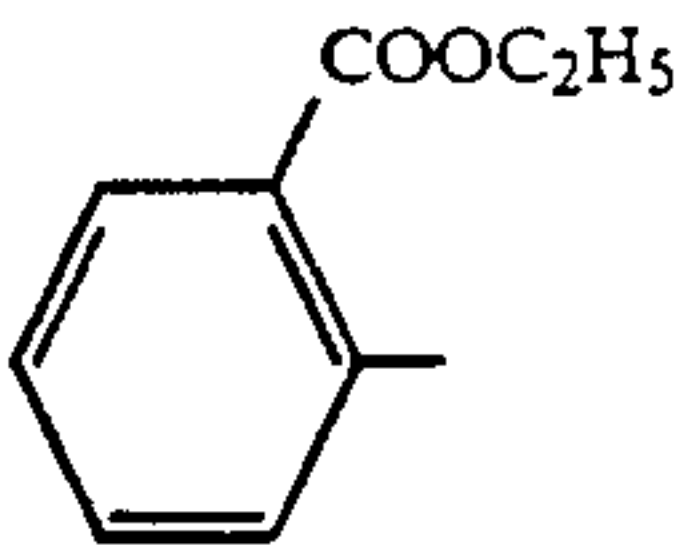
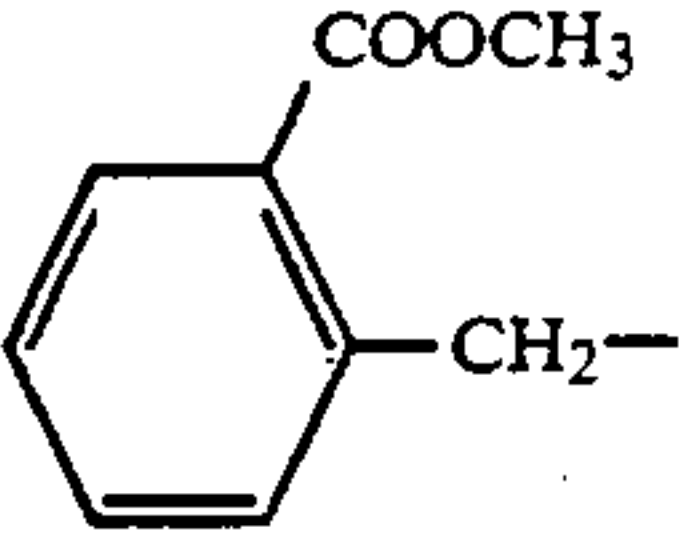
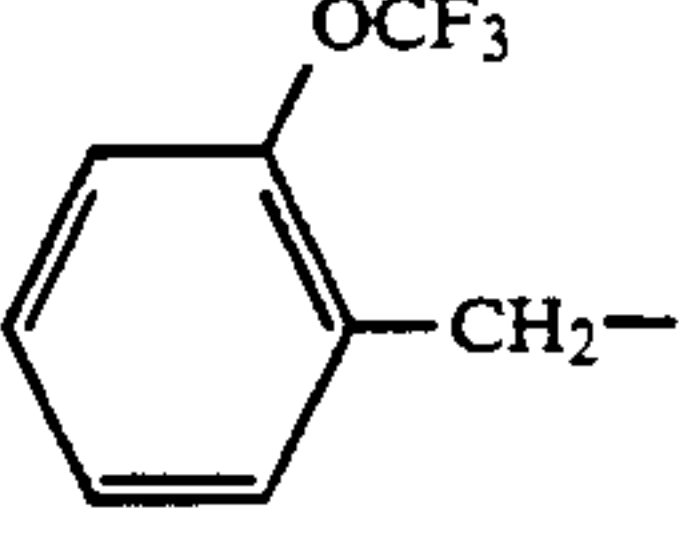
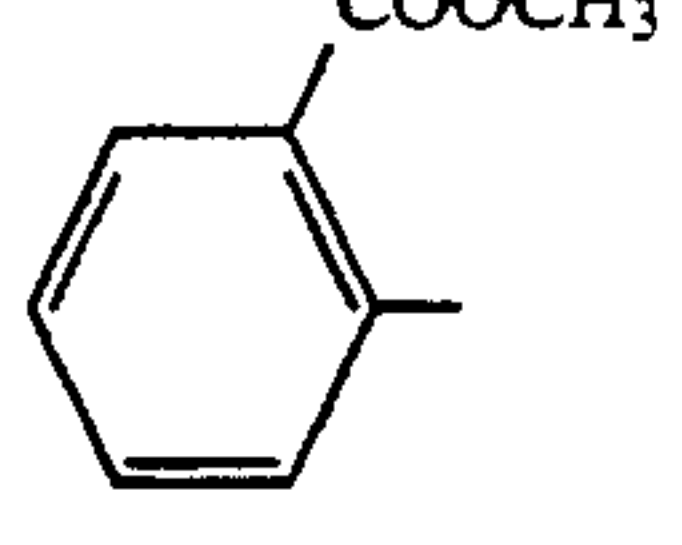
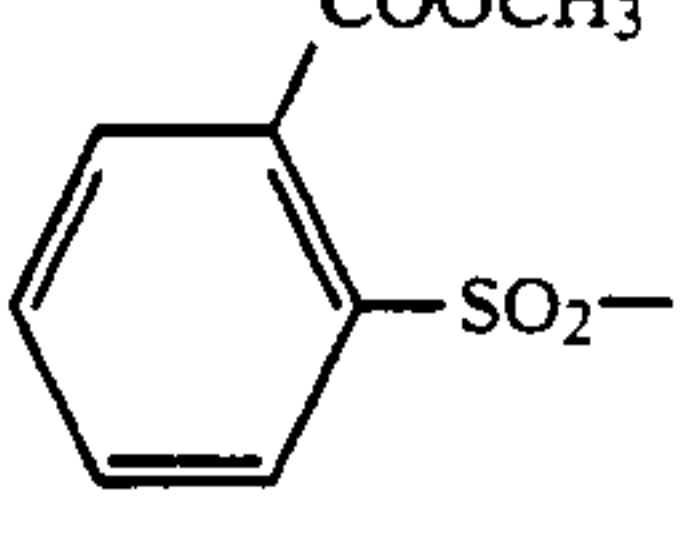
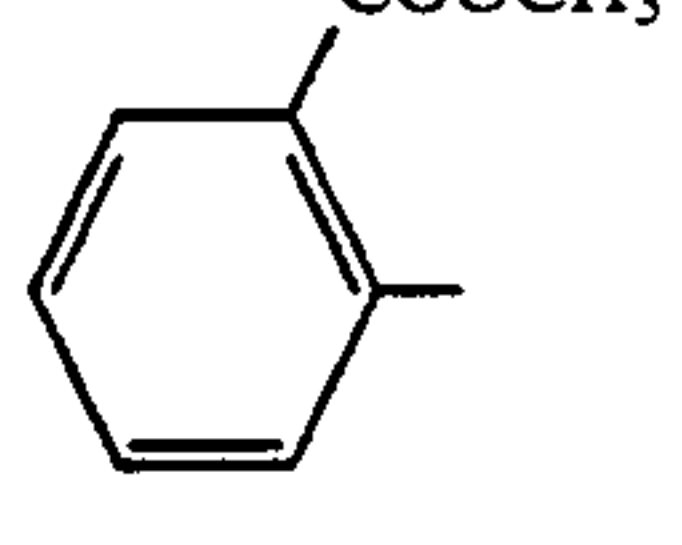
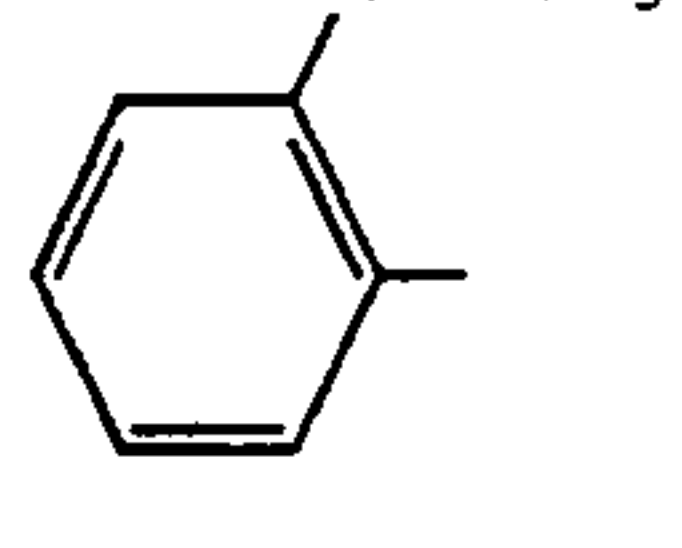
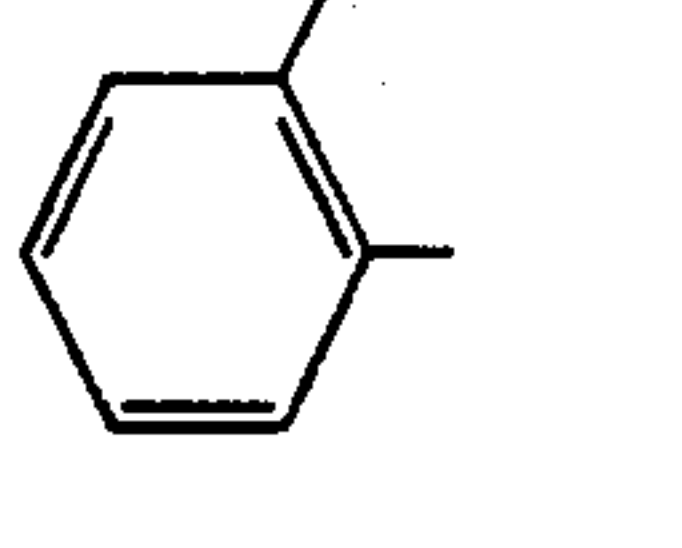
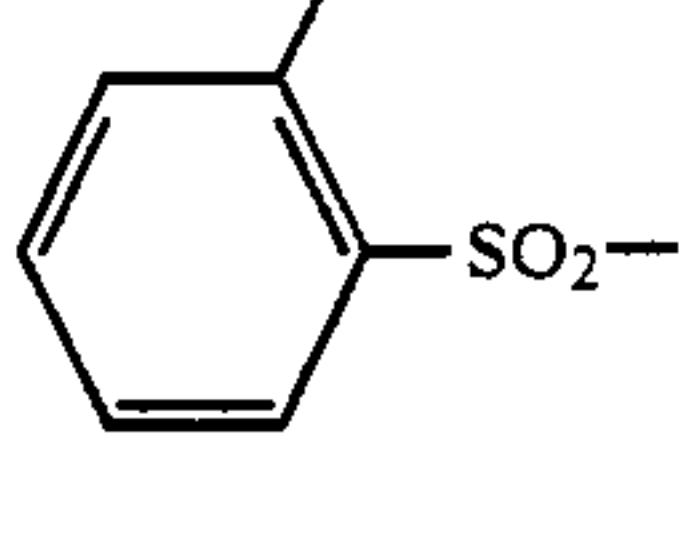
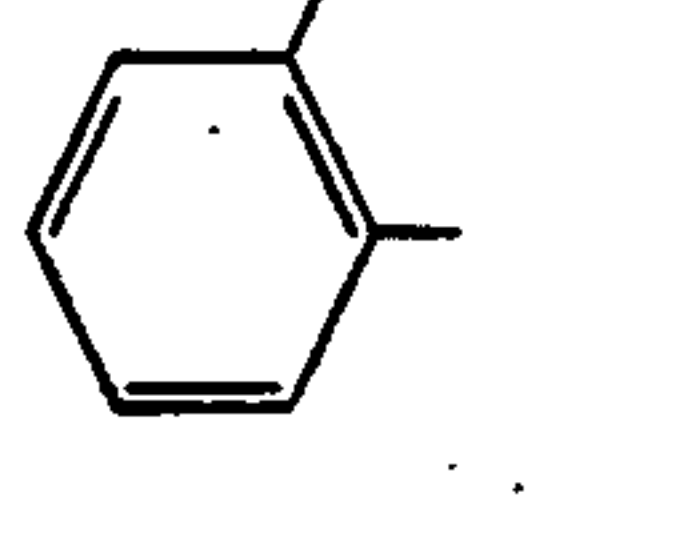
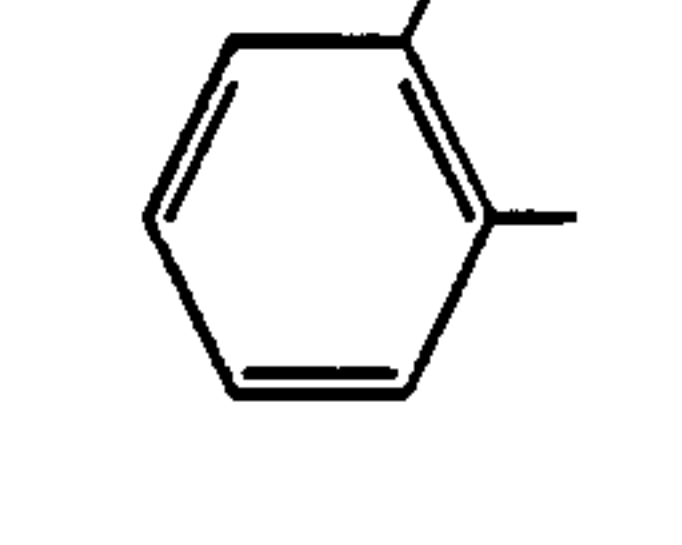
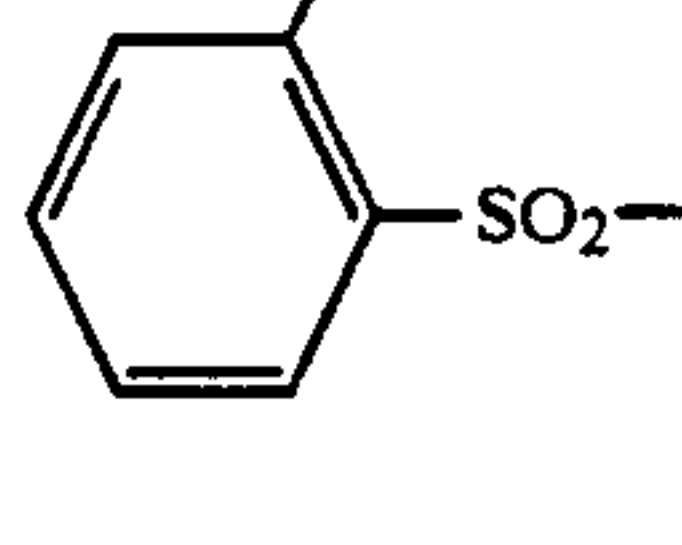
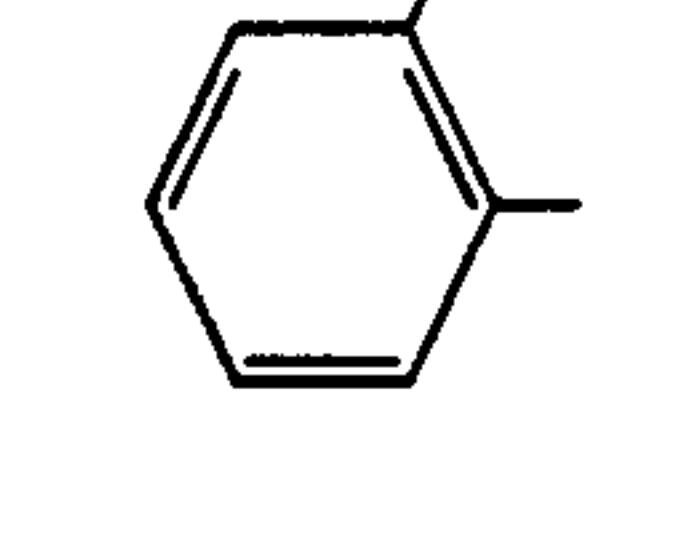
Examples of the compounds of the formula (I)								
Ex- am- ple No.	R ¹	R ²	R ³	R ⁴	X	Y	Z	Melting point (°C.)
6		H	CH ₃	SO ₂ CH ₃	N	CH	C—OCH ₃	140
7		H	CH ₃	OCH ₃	N	CH	C—OCH ₃	178
8		H	CH ₃	OCH ₃	N	CH	C—OCH ₃	186
9			H	CH ₃	N	CH	C—CH ₃	
10		H	H	CH ₃	N	CH	C—CH ₃	
11		Na	H	CH ₃	N	CH	C—CH ₃	
12			H	CH ₃	N	CH	C—CH ₃	
13		H	H	CH ₃	N	CH	C—CH ₃	
14			H	CH ₃	N	CH	C—CH ₃	
15		H	H	CH ₃	N	CH	C—CH ₃	

TABLE 2-continued

Examples of the compounds of the formula (I)								Melting point (°C.)
Ex-ample No.	R ¹	R ²	R ³	R ⁴	X	Y	Z	
16			H	CH ₃	N	CH	C-CH ₃	
17		H	H	CH ₃	N	CH	C-CH ₃	
18			H	CH ₃	N	CH	C-CH ₃	
19		H	H	CH ₃	N	CH	C-CH ₃	
20			H	CH ₃	N	CH	C-CH ₃	
21			H	CH ₃	N	N	C-CH ₃	
22		H	H	CH ₃	N	CH	C-CH ₃	
23			H	CH ₃	N	CH	C-CH ₃	
24			H	CH ₃	N	CH	C-CH ₃	
25		H	H	CH ₃	N	CH	C-CH ₃	

TABLE 2-continued

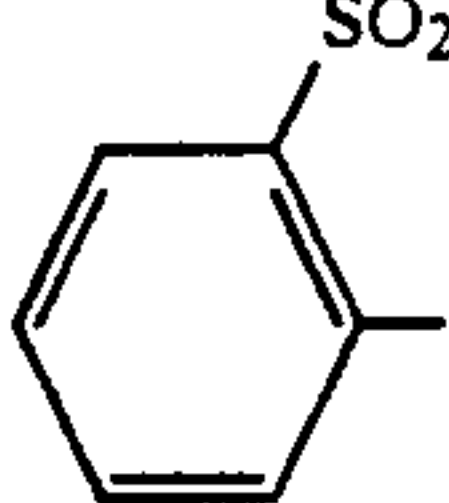
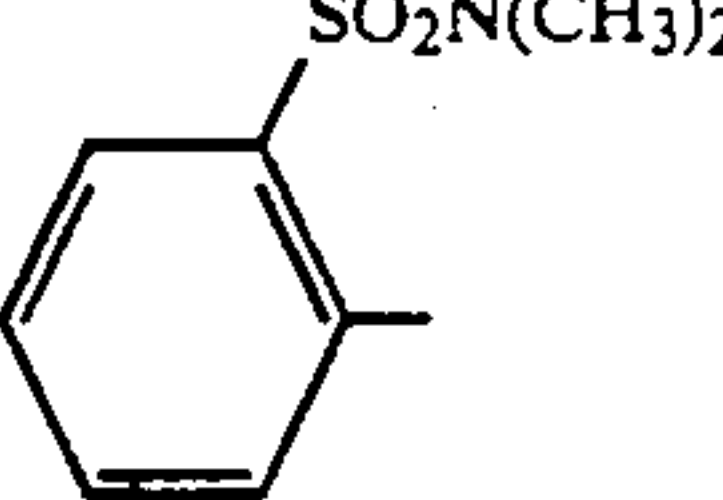
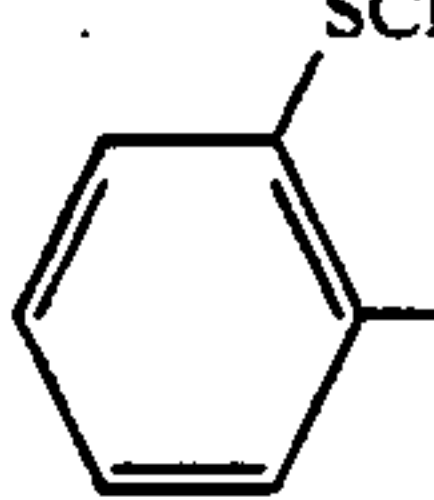
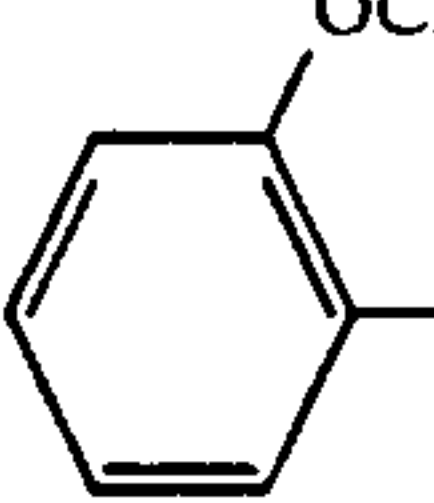
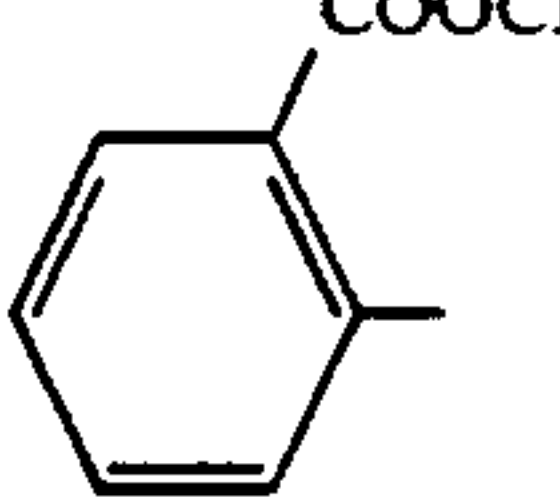
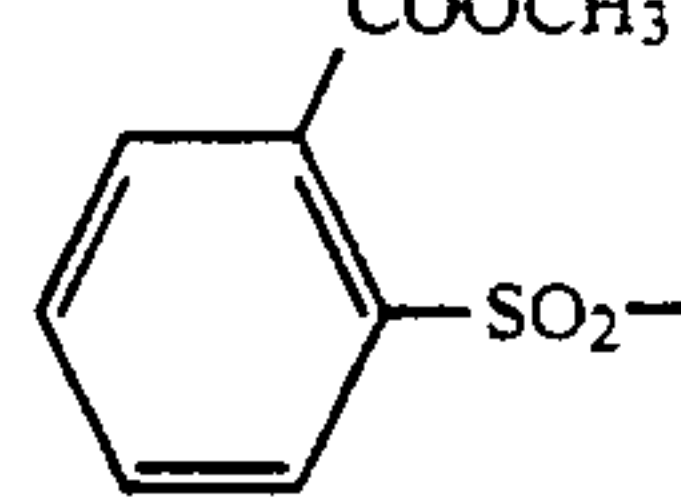
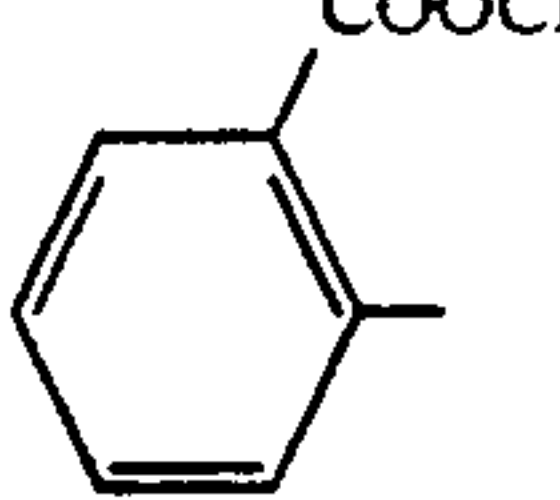
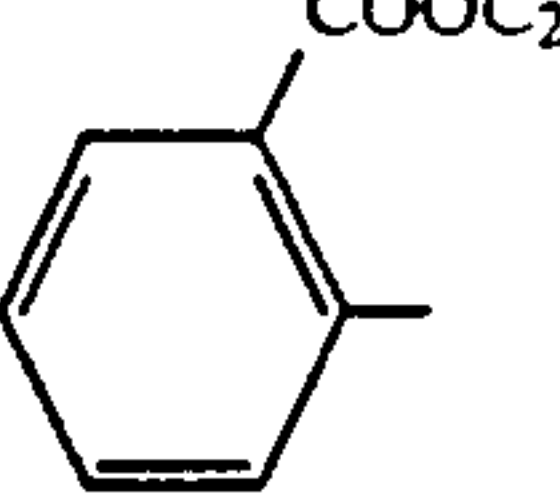
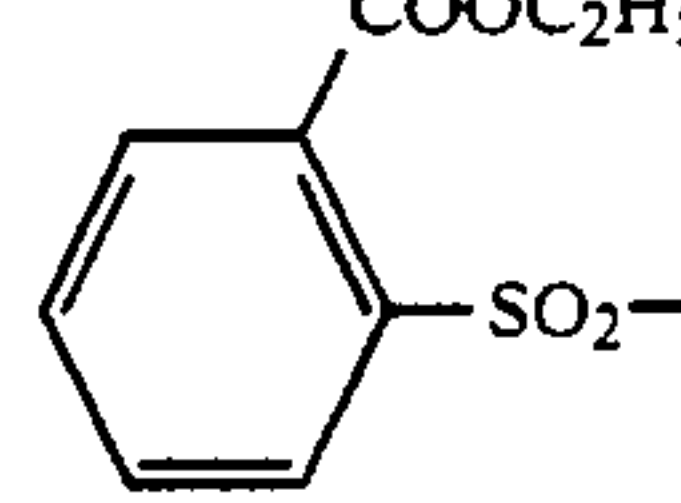
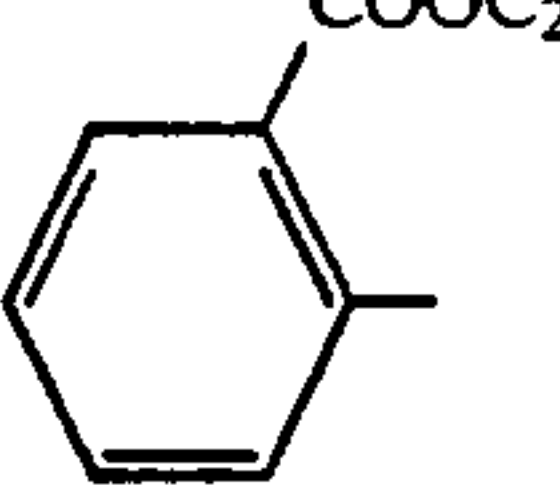
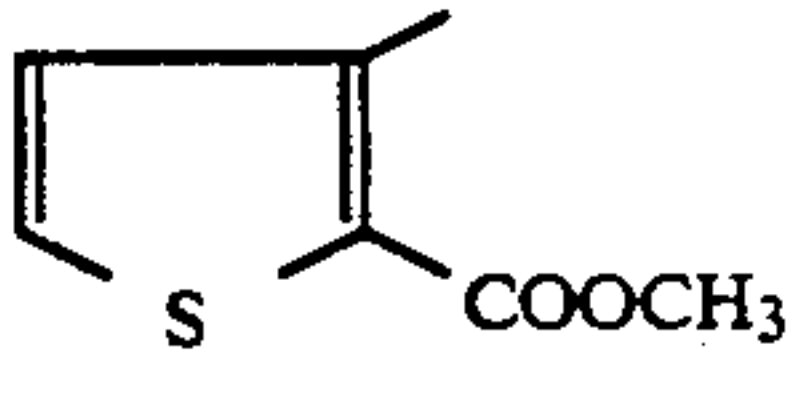
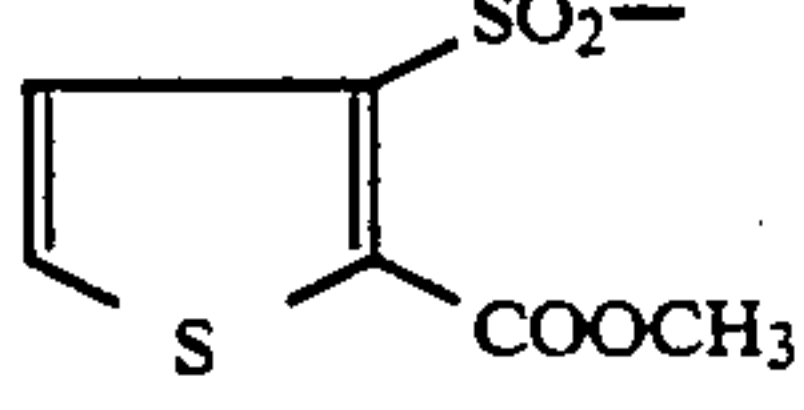
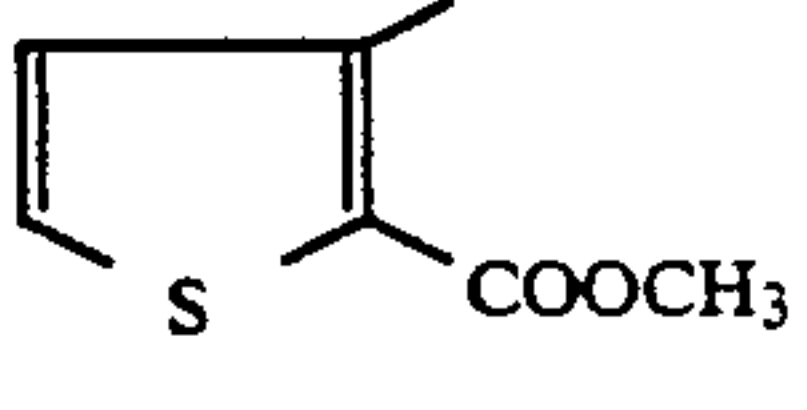
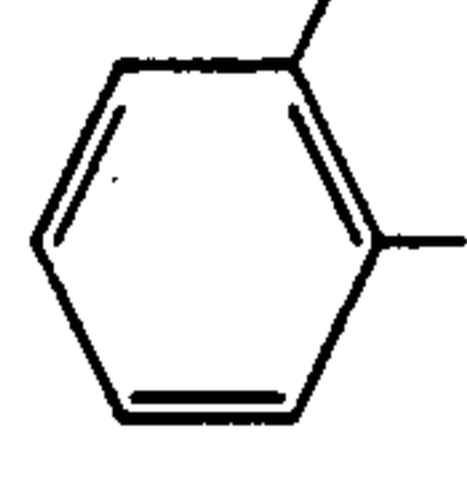
Ex- am- ple No.	Examples of the compounds of the formula (I)							Melting point (°C.)
	R ¹	R ²	R ³	R ⁴	X	Y	Z	
26		H	H	CH ₃	N	CH	C-CH ₃	
27		H	H	CH ₃	N	CH	C-CH ₃	
28		H	H	CH ₃	N	CH	C-CH ₃	
29		H	H	CH ₃	N	CH	C-CH ₃	
30			H	CH ₃	N	CH	C-OCH ₃	
31		H	H	CH ₃	N	CH	C-OCH ₃	
32			H	CH ₃	N	CH	C-OCH ₃	
33		H	H	CH ₃	N	CH	C-OCH ₃	
34			H	CH ₃	N	CH	C-OCH ₃	
35		H	H	CH ₃	N	CH	C-OCH ₃	
36		H	H	CH ₃	N	CH	C-OCH ₃	181

TABLE 2-continued

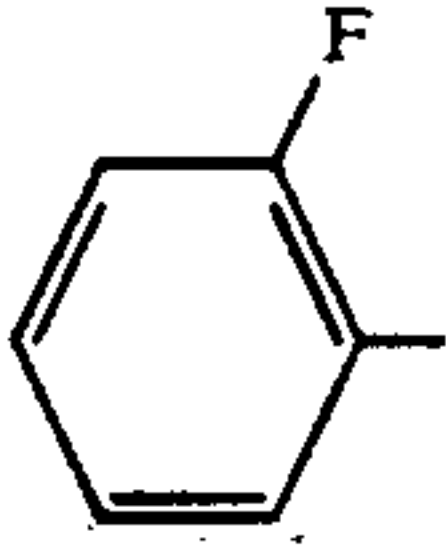
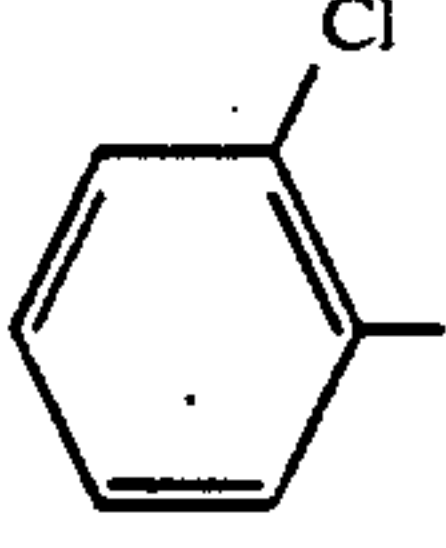
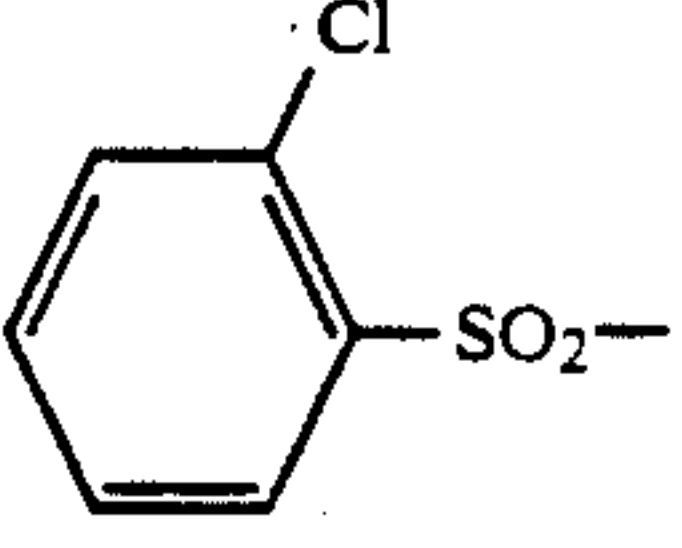
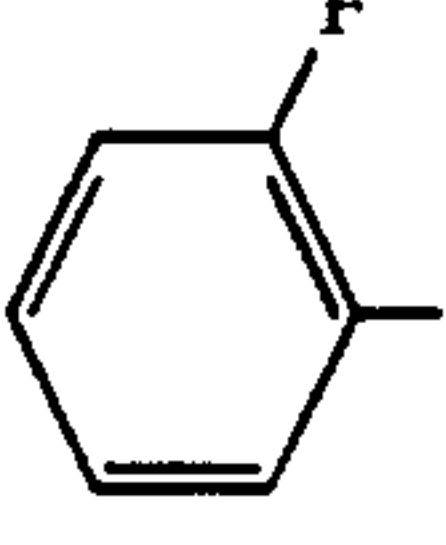
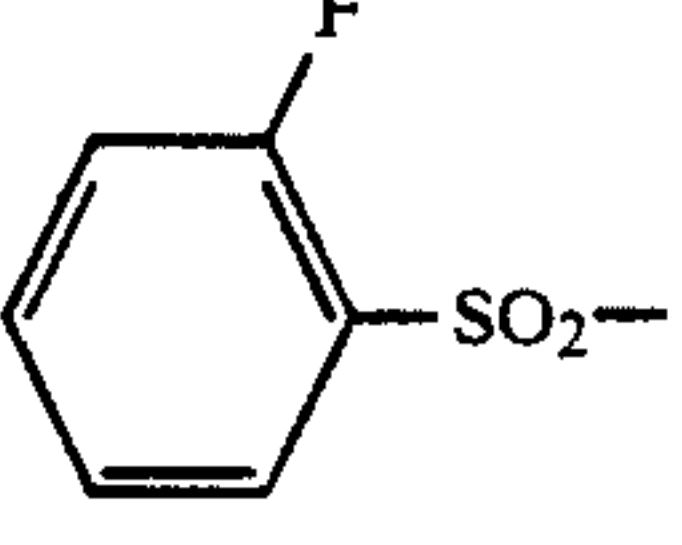
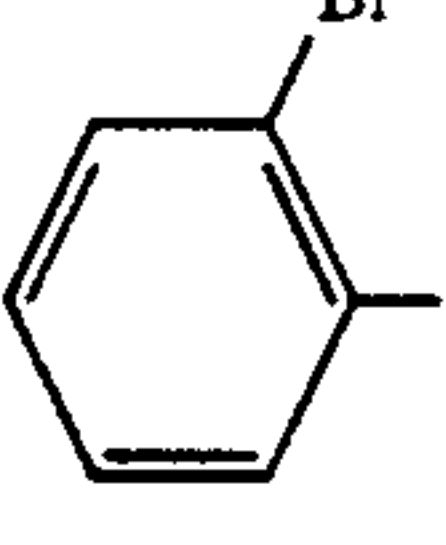
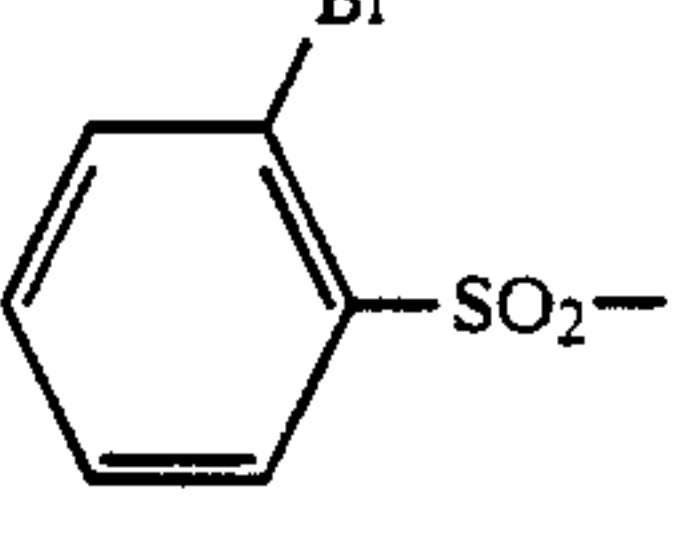
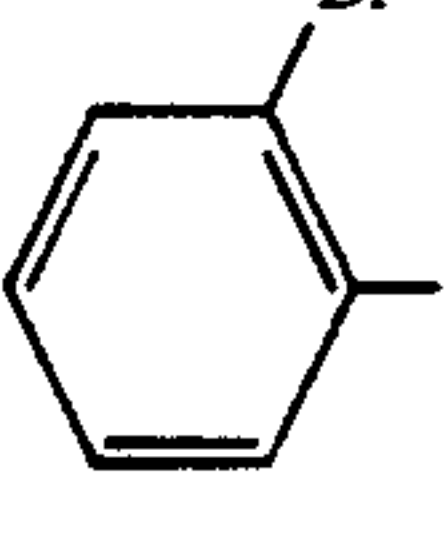
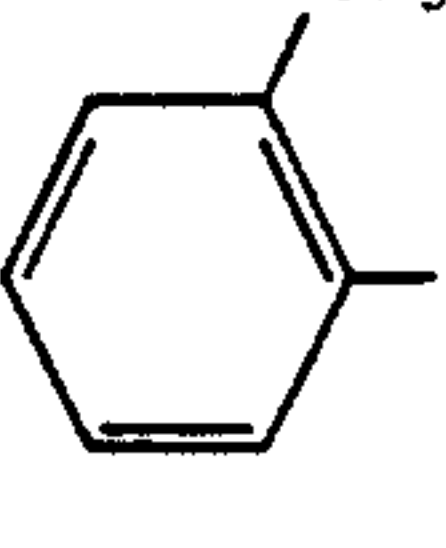
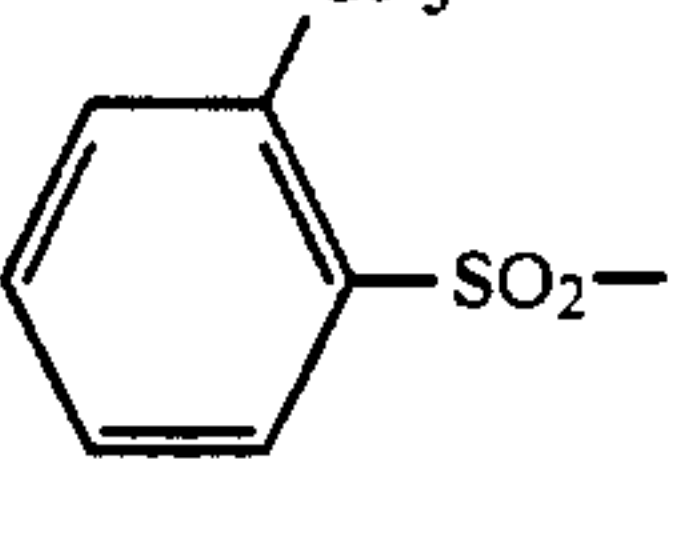
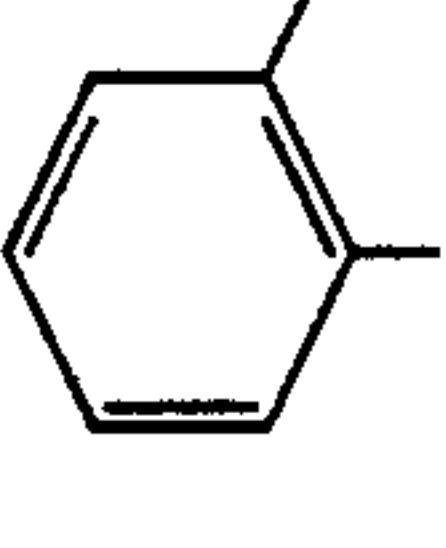
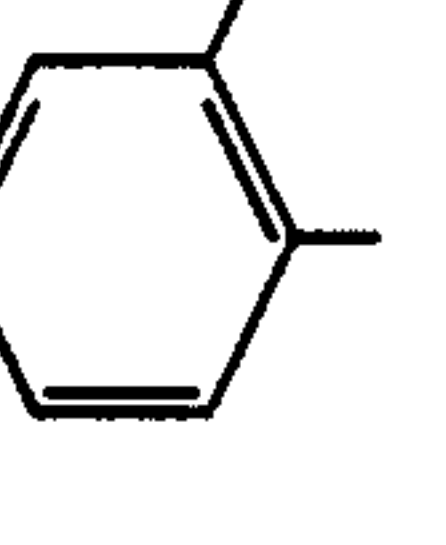
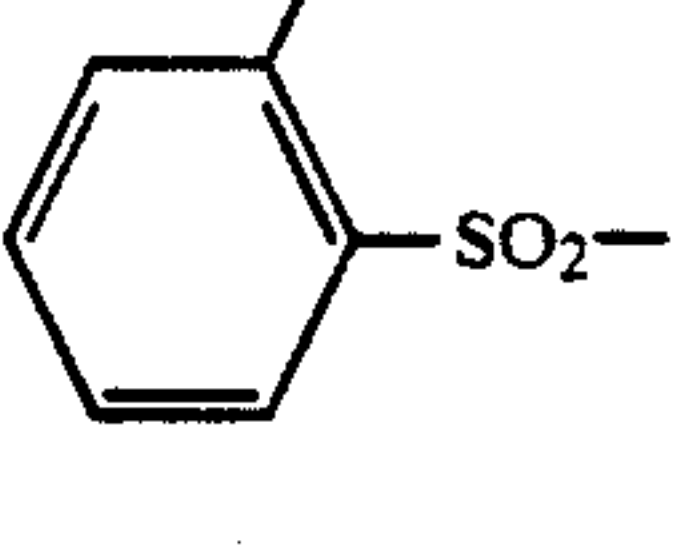
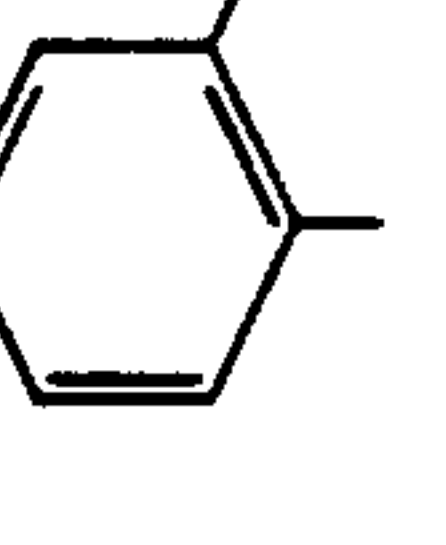
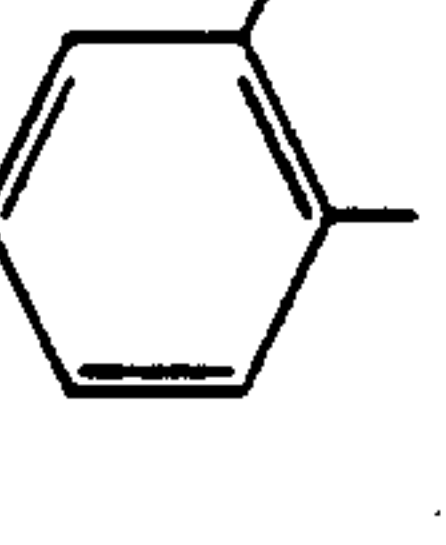
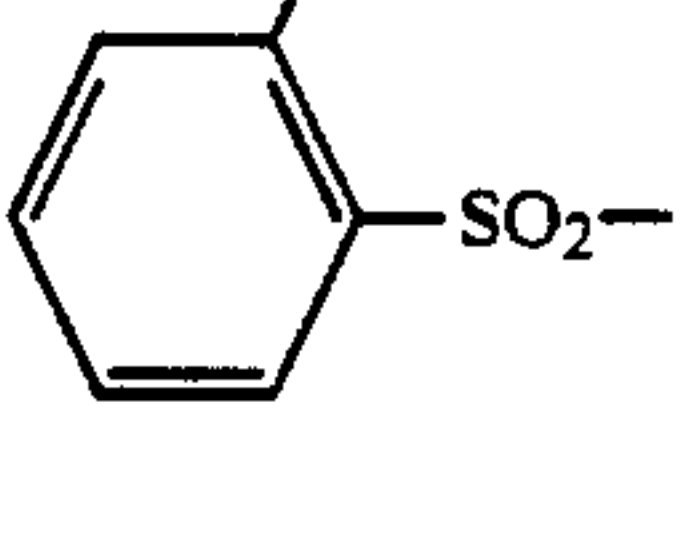
Ex- am- ple No.	Examples of the compounds of the formula (I)							Melting point (°C.)
	R ¹	R ²	R ³	R ⁴	X	Y	Z	
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38			H	CH ₃	N	CH	C—OCH ₃	
39			H	CH ₃	N	CH	C—OCH ₃	
40			H	CH ₃	N	CH	C—OCH ₃	
41		H	H	CH ₃	N	CH	C—OCH ₃	
42			H	CH ₃	N	CH	C—OCH ₃	
43		H	H	CH ₃	N	CH	C—OCH ₃	
44			H	CH ₃	N	CH	C—OCH ₃	
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TABLE 2-continued

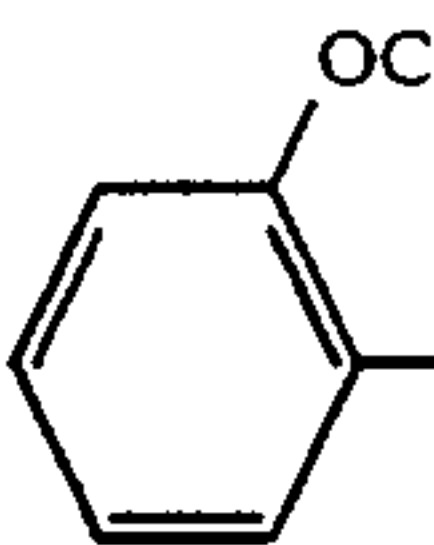
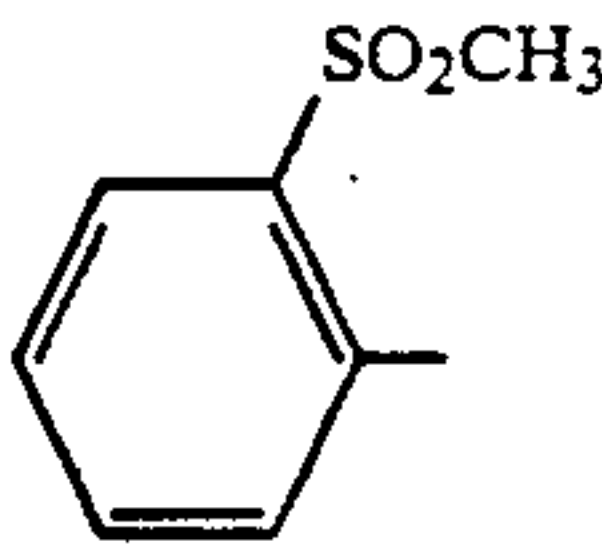
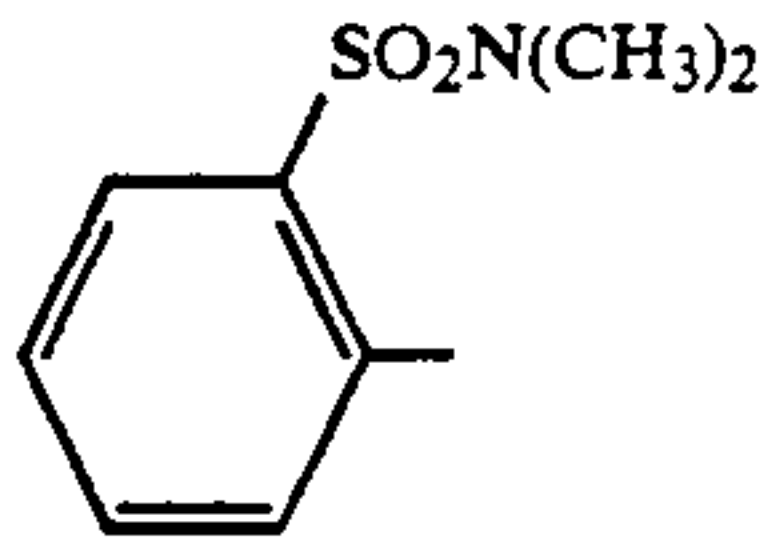
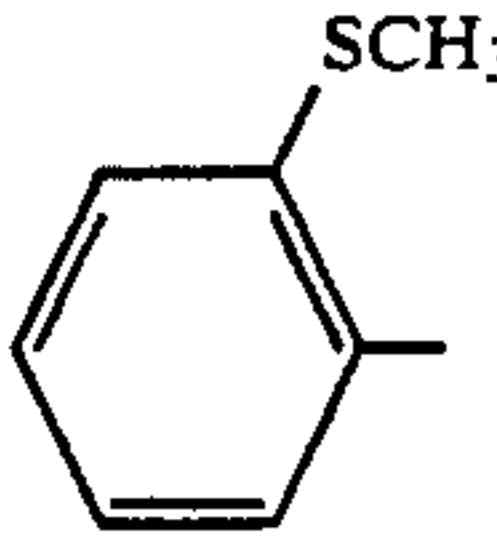
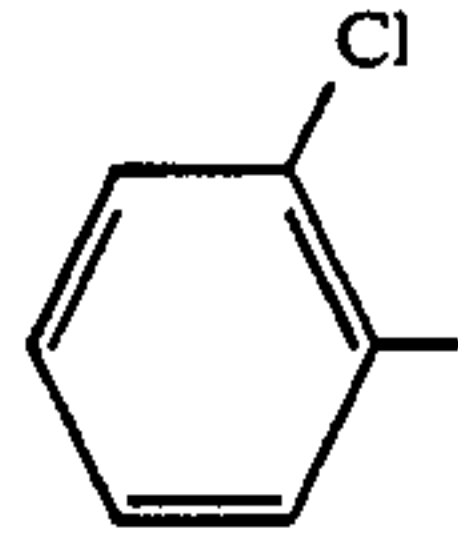
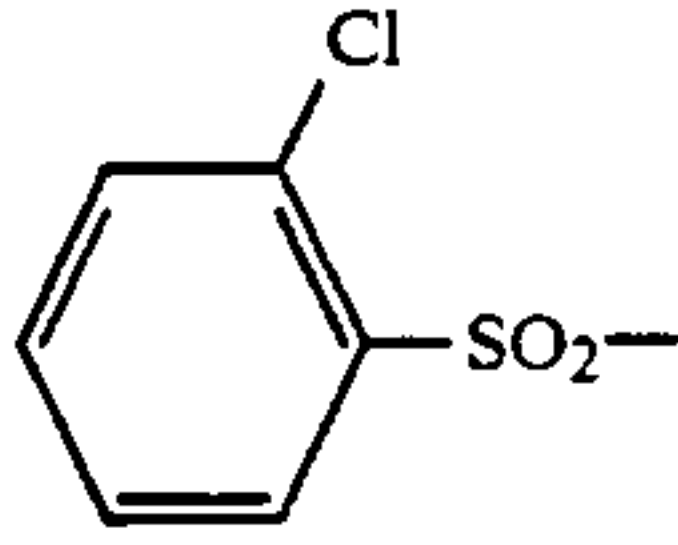
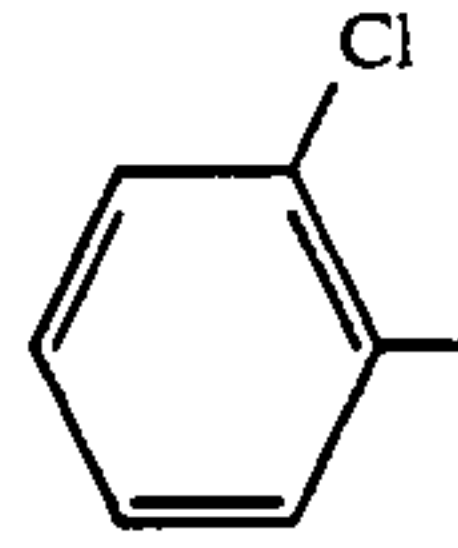
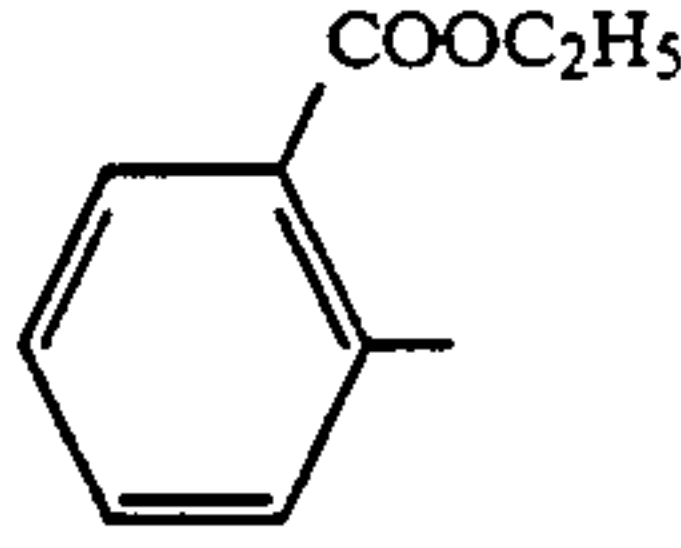
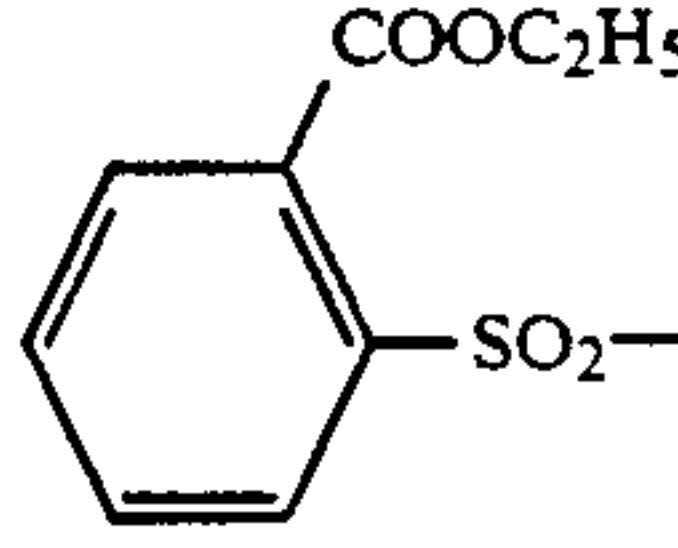
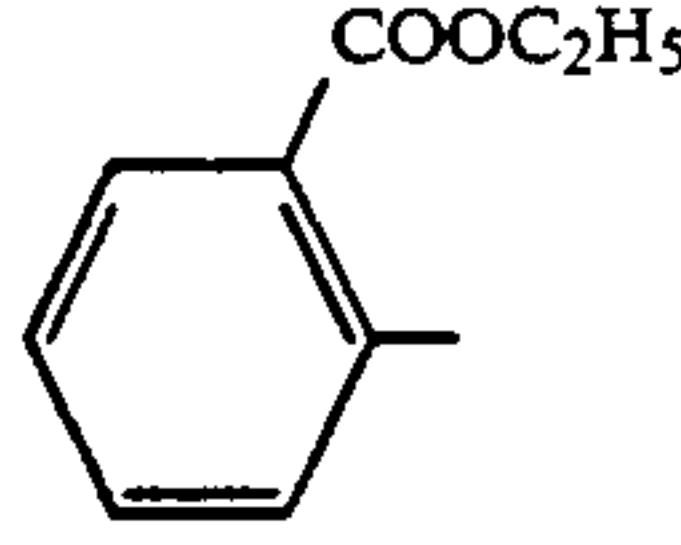
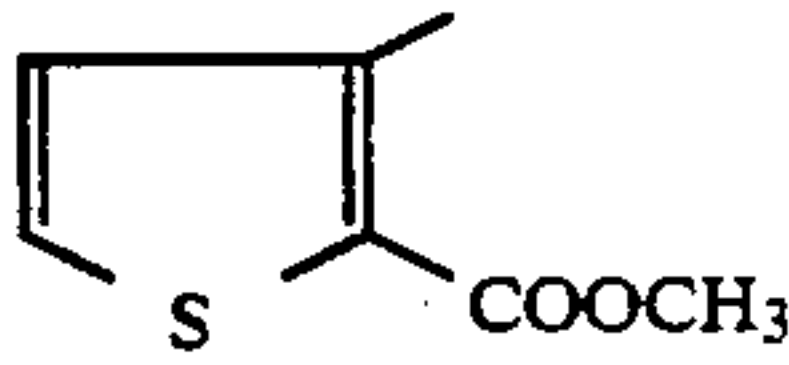
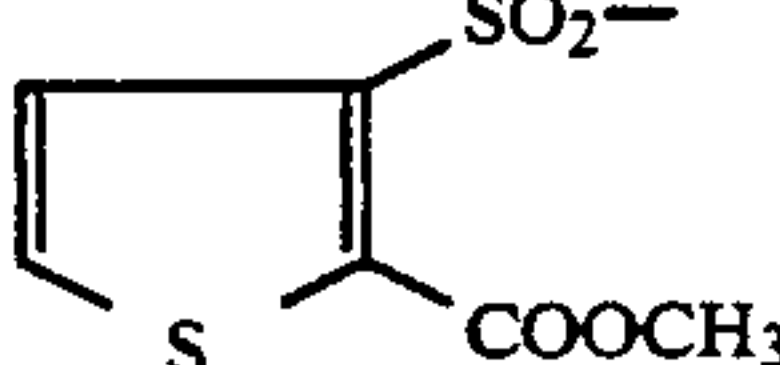
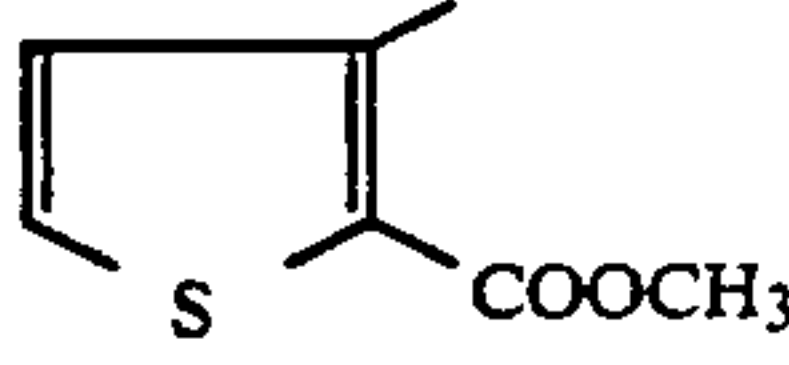
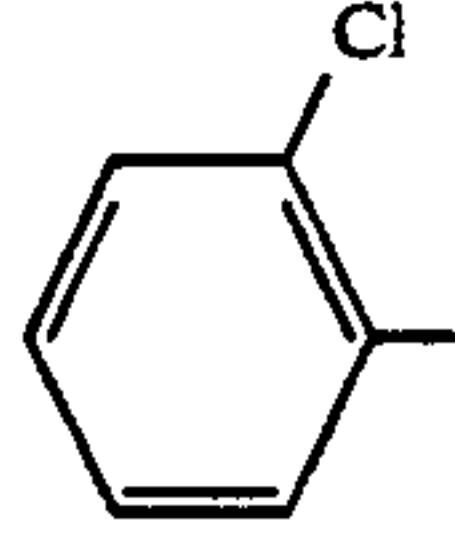
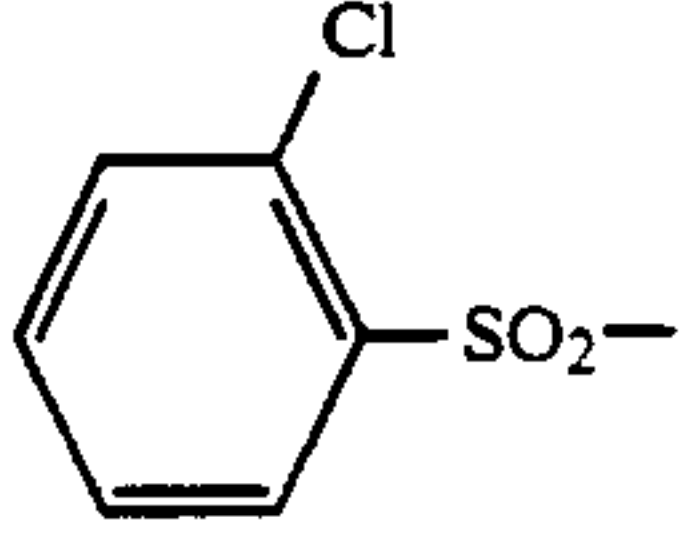
Ex- am- ple No.	Examples of the compounds of the formula (I)							Melting point (°C.)
	R ¹	R ²	R ³	R ⁴	X	Y	Z	
47		H	H	CH ₃	N	CH	C—OCH ₃	219
48		H	H	CH ₃	N	CH	C—OCH ₃	
49		H	H	CH ₃	N	CH	C—OCH ₃	
50		H	H	CH ₃	N	CH	C—OCH ₃	
51			H	CH ₃	N	CH	C—CH ₃	
52		H	H	CH ₃	N	CH	C—CH ₃	
53			H	OCH ₃	N	CH	C—OCH ₃	
54		H	H	OCH ₃	N	CH	C—OCH ₃	
55			H	OCH ₃	N	CH	C—OCH ₃	
56		H	H	OCH ₃	N	CH	C—OCH ₃	
57			H	OCH ₃	N	CH	C—OCH ₃	

TABLE 2-continued

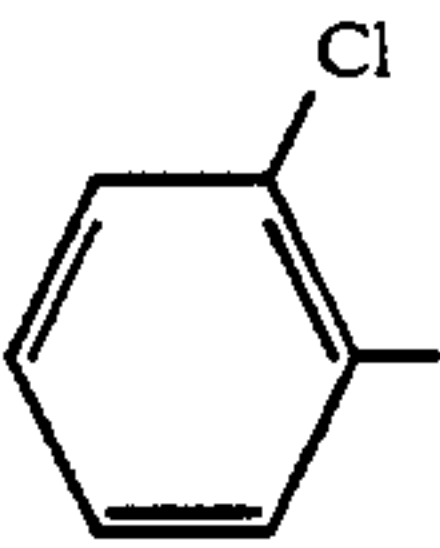
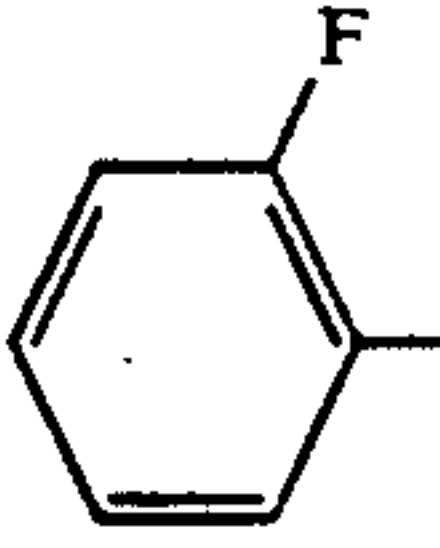
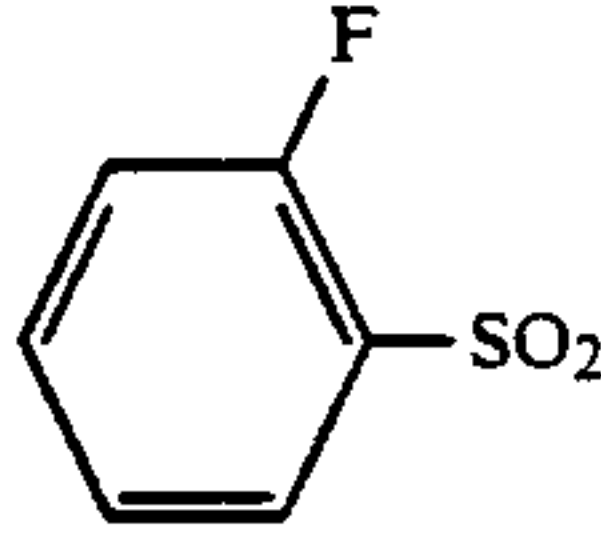
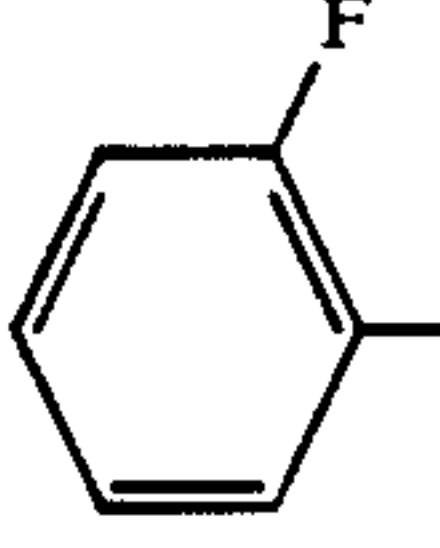
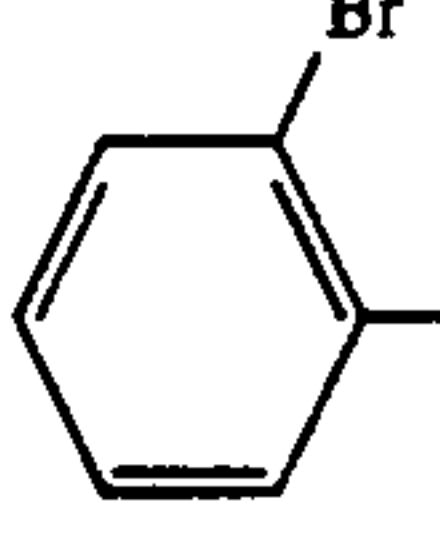
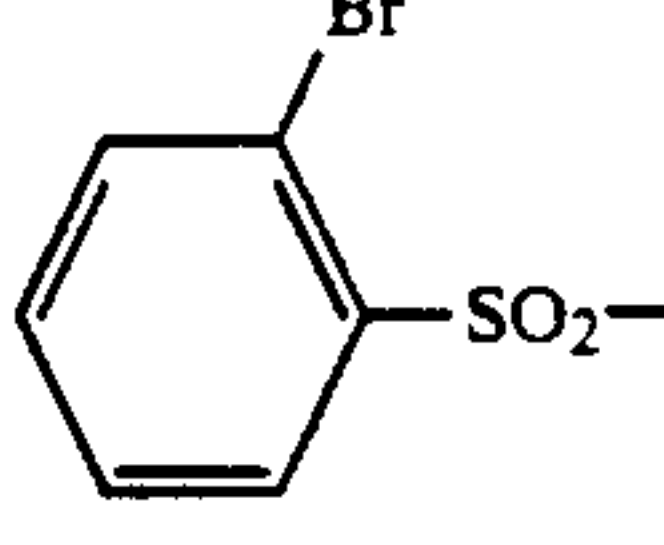
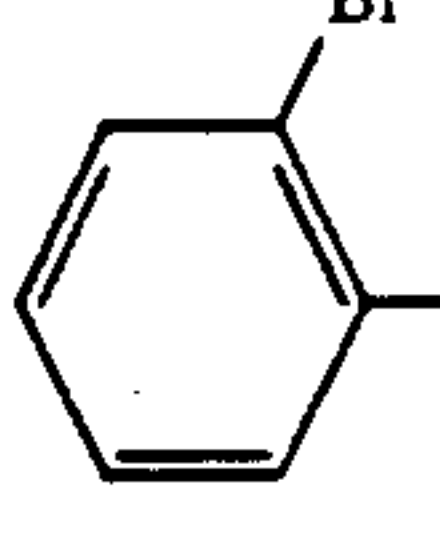
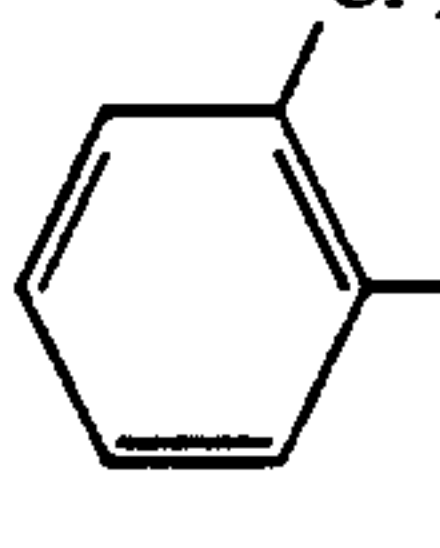
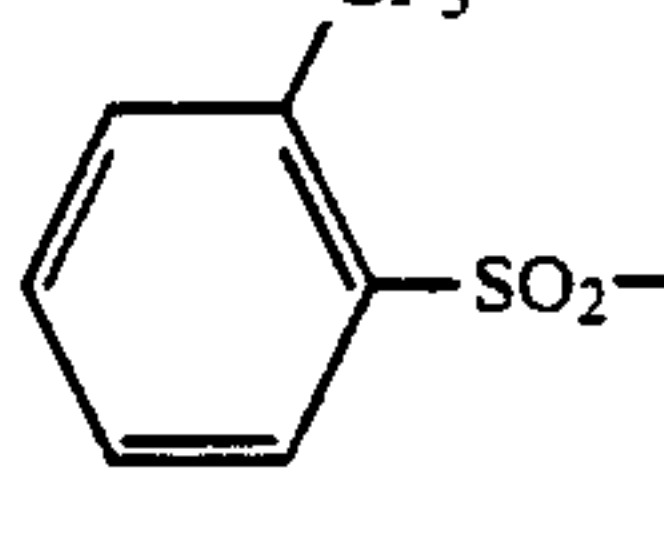
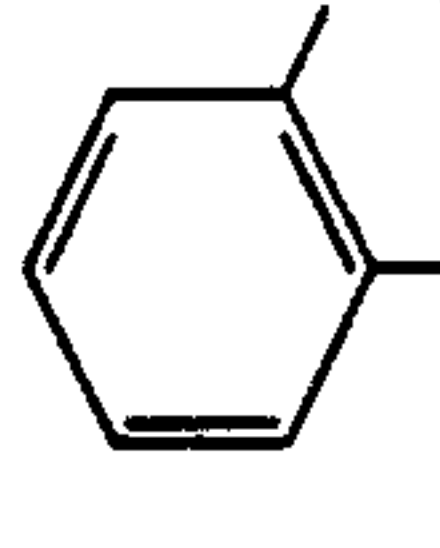
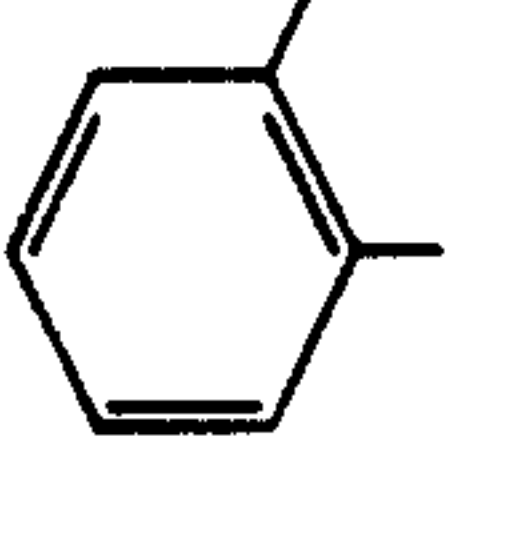
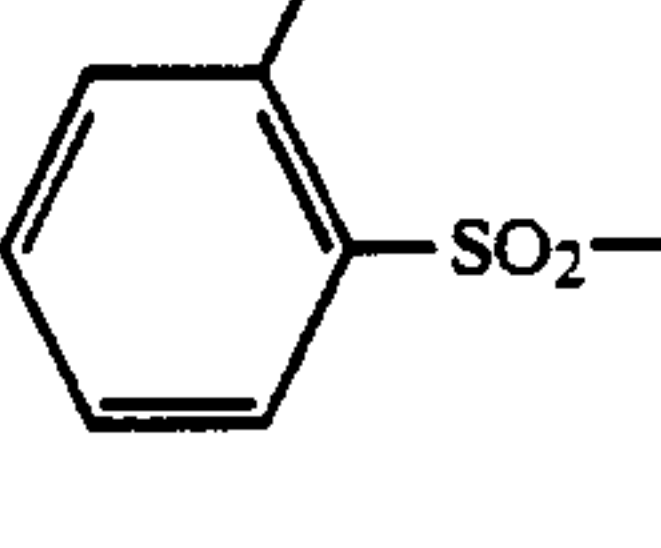
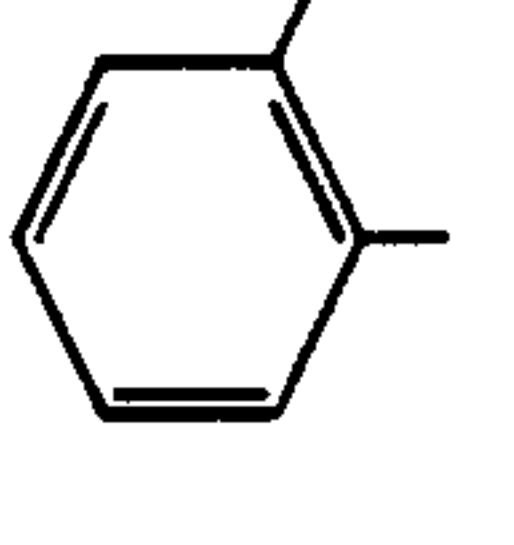
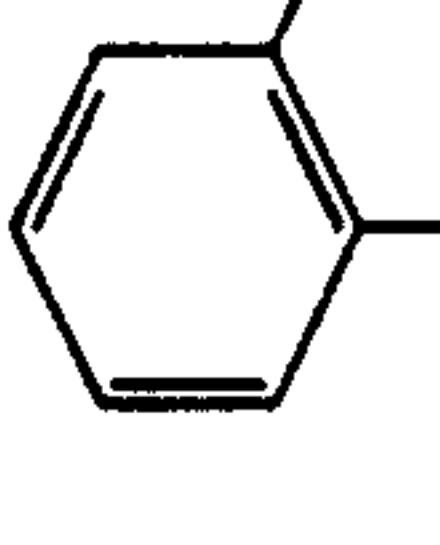
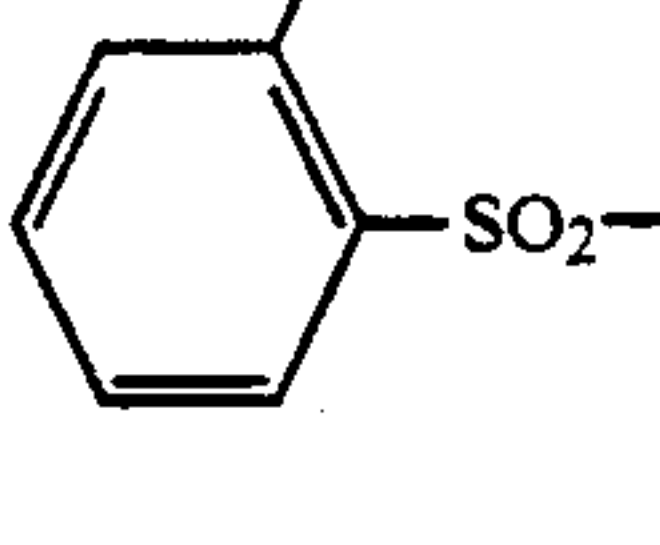
Examples of the compounds of the formula (I)								Melting point (°C.)
Ex-ample No.	R ¹	R ²	R ³	R ⁴	X	Y	Z	
58		H	H	OCH ₃	N	CH	C—OCH ₃	
59			H	OCH ₃	N	CH	C—OCH ₃	
60		H	H	OCH ₃	N	CH	C—OCH ₃	
61			H	OCH ₃	N	CH	C—OCH ₃	
62		H	H	OCH ₃	N	CH	C—OCH ₃	
63			H	OCH ₃	N	CH	C—OCH ₃	
64		H	H	OCH ₃	N	CH	C—OCH ₃	
65			H	OCH ₃	N	CH	C—OCH ₃	
66		H	H	OCH ₃	N	CH	C—OCH ₃	
67			H	OCH ₃	N	CH	C—OCH ₃	

TABLE 2-continued

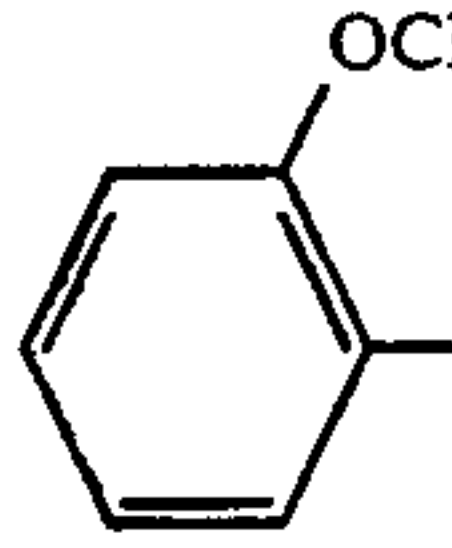
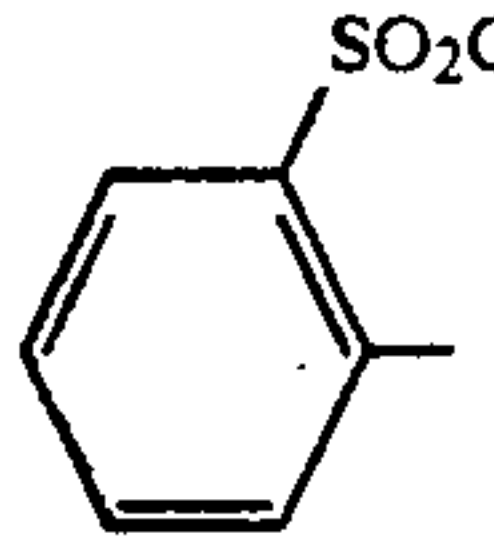
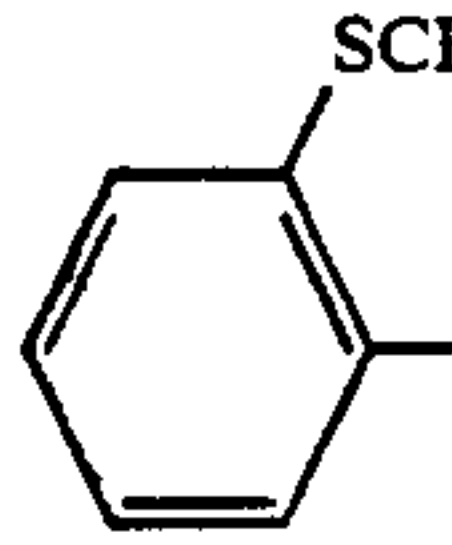
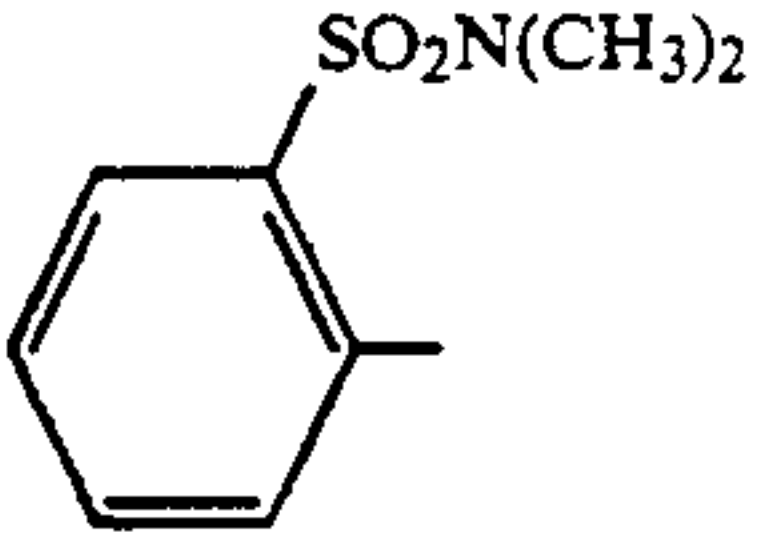
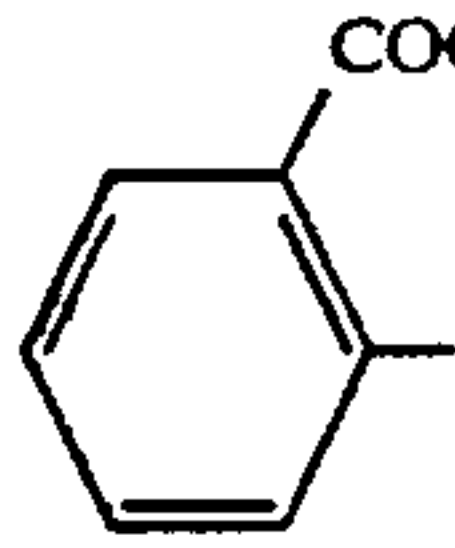
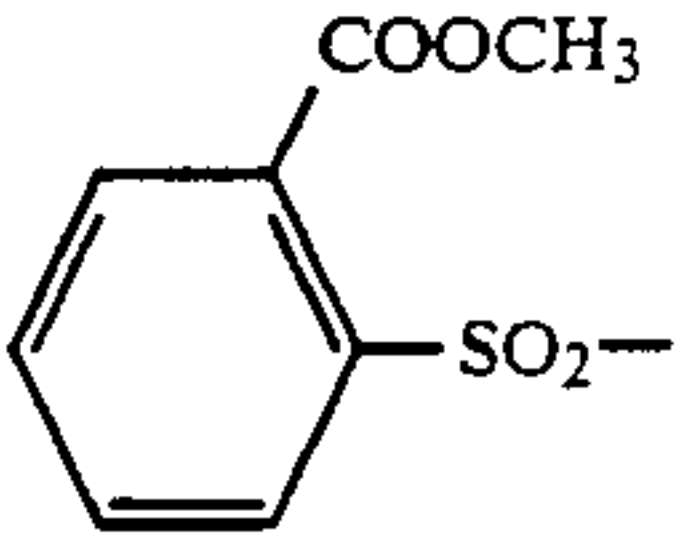
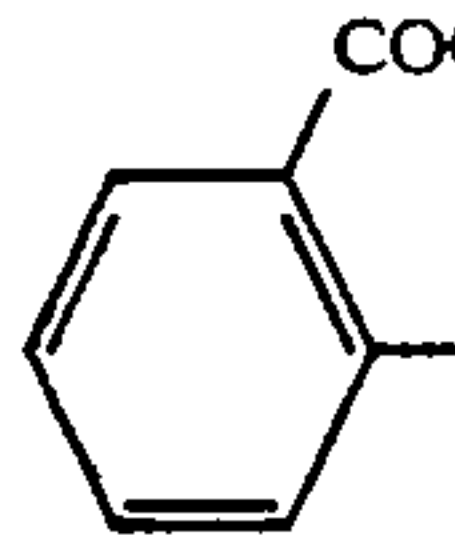
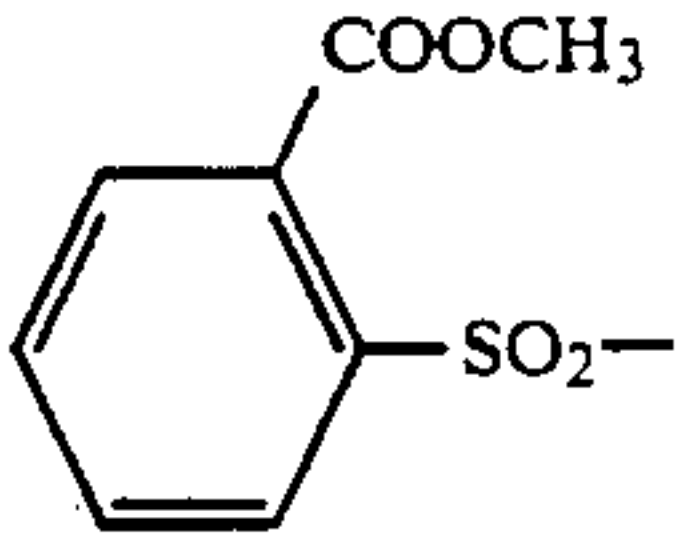
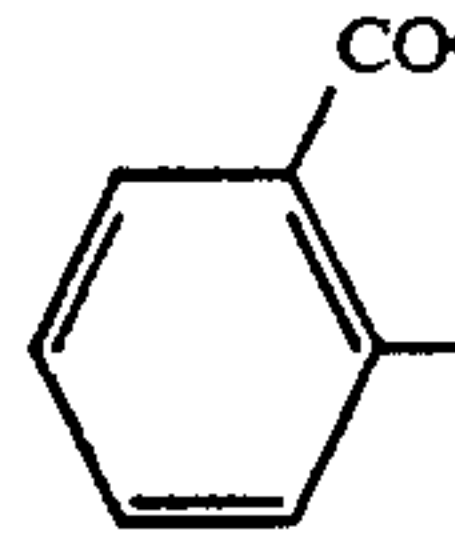
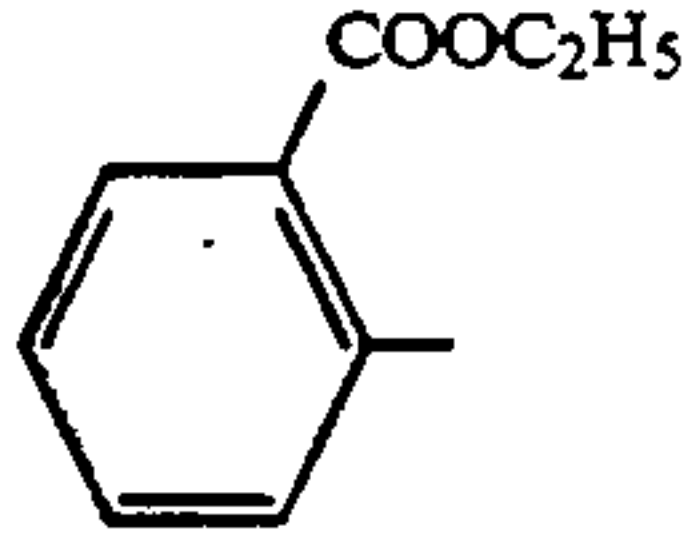
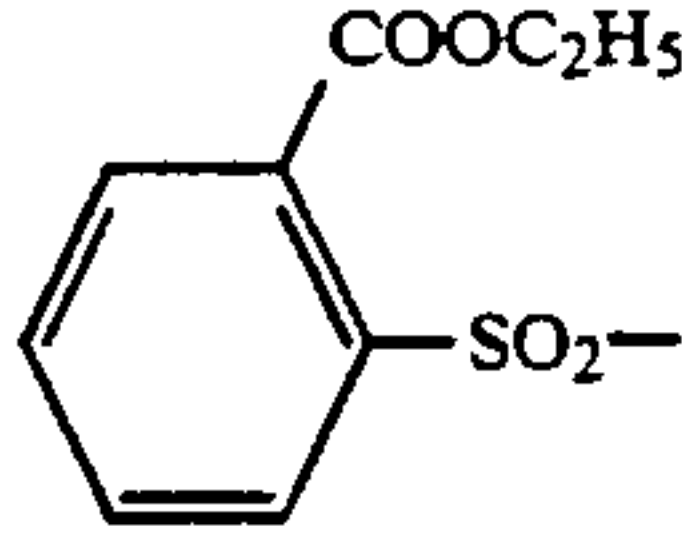
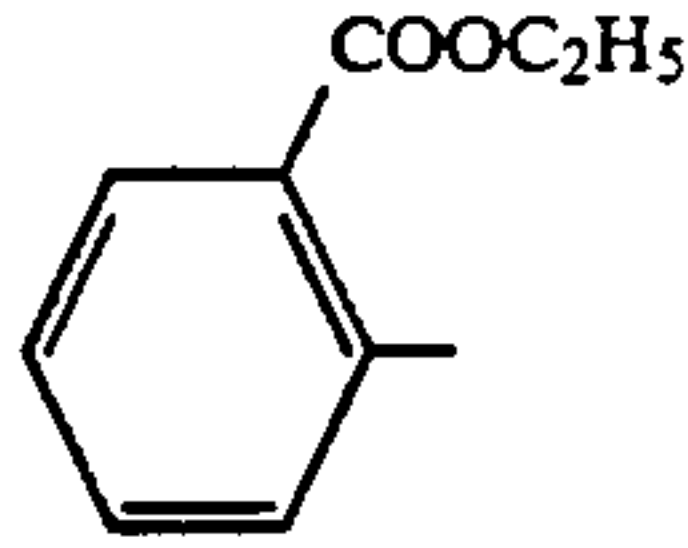
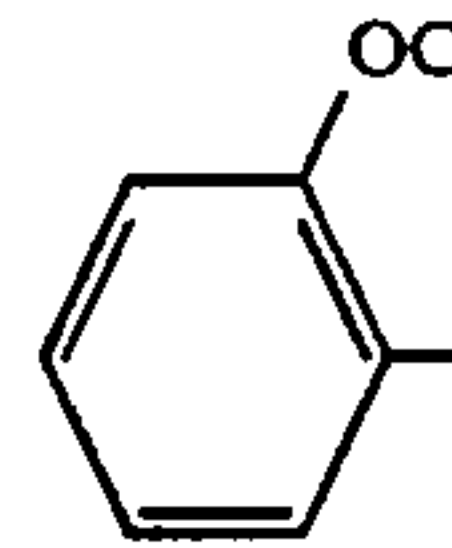
Ex- am- ple No.	Examples of the compounds of the formula (I)							Melting point (°C.)
	R ¹	R ²	R ³	R ⁴	X	Y	Z	
68		H	H	OCH ₃	N	CH	C—OCH ₃	208
69		H	H	OCH ₃	N	CH	C—OCH ₃	
70		H	H	OCH ₃	N	CH	C—OCH ₃	
71		H	H	OCH ₃	N	CH	C—OCH ₃	
72			H	CF ₃	N	CH	C—OCH ₃	
73			H	OCH ₃	N	CH	C—Cl	
74		H	H	CF ₃	N	CH	C—OCH ₃	
75			H	CF ₃	N	CH	C—OCH ₃	
76		H	H	CF ₃	N	CH	C—OCH ₃	
77		H	H	CF ₃	N	CH	C—OCH ₃	

TABLE 2-continued

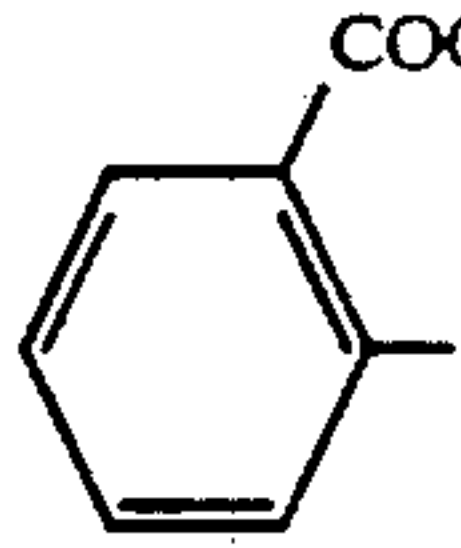
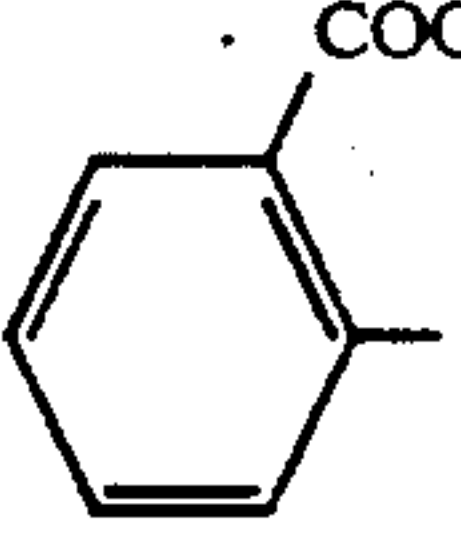
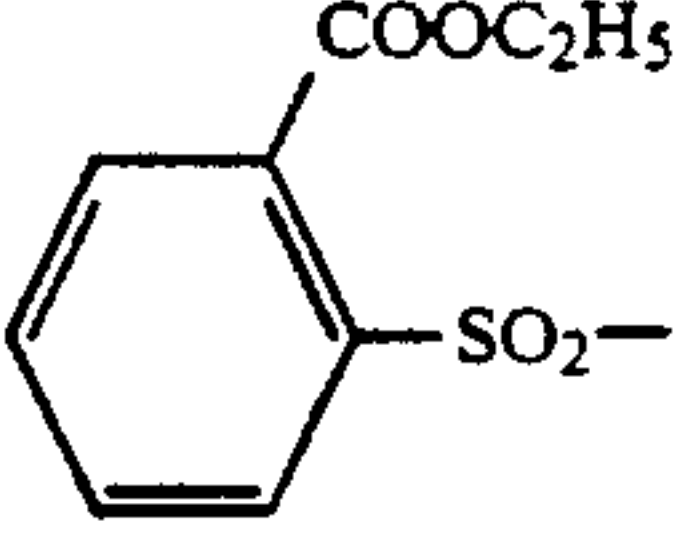
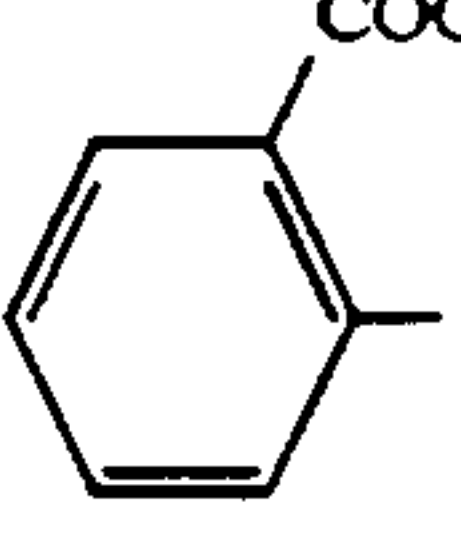
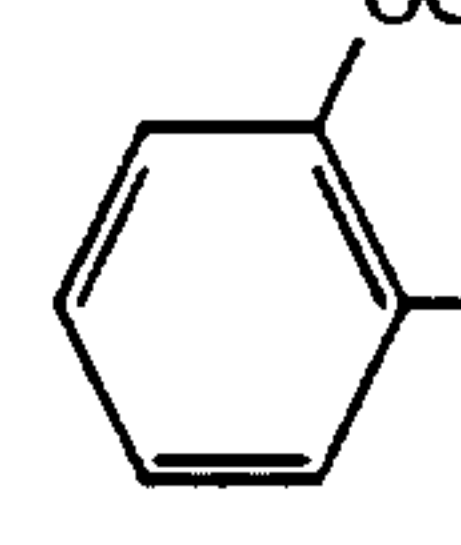
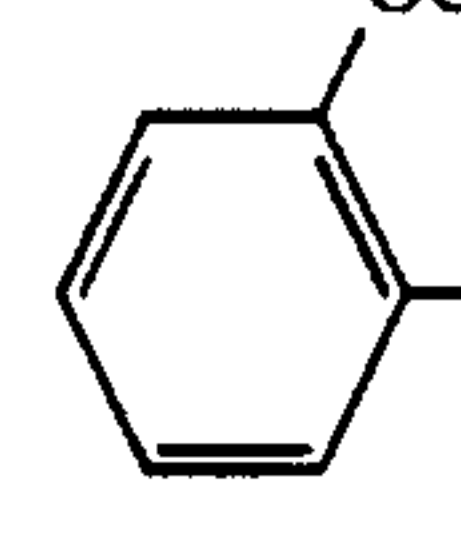
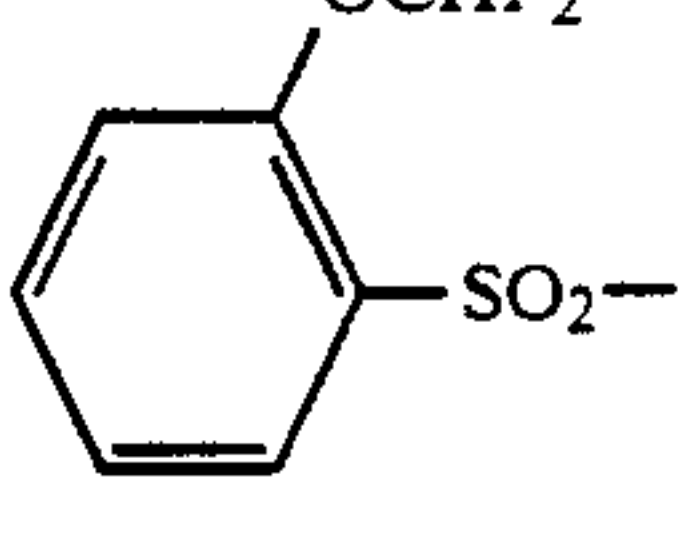
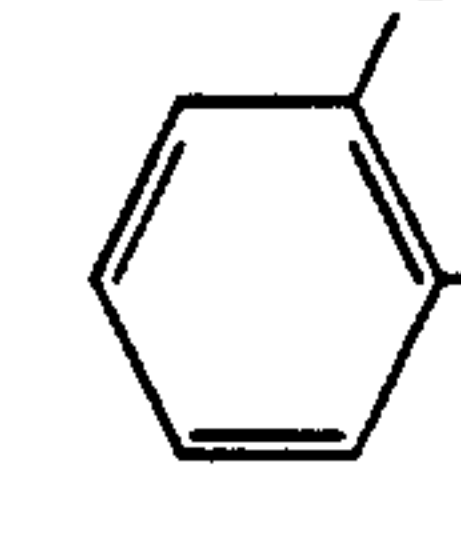
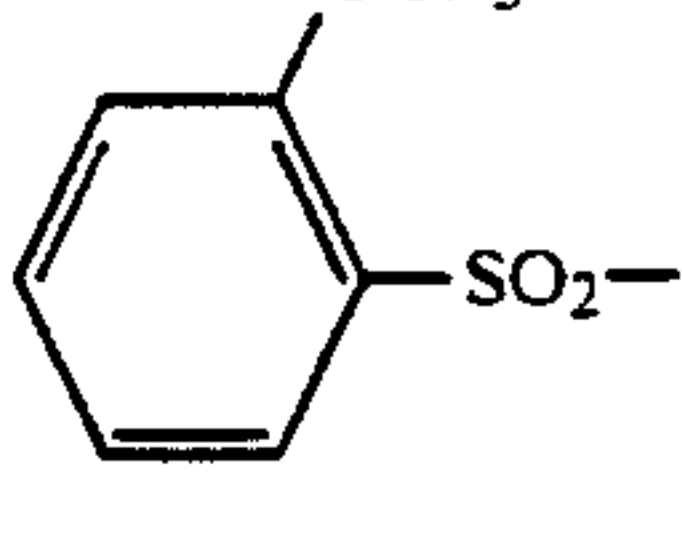
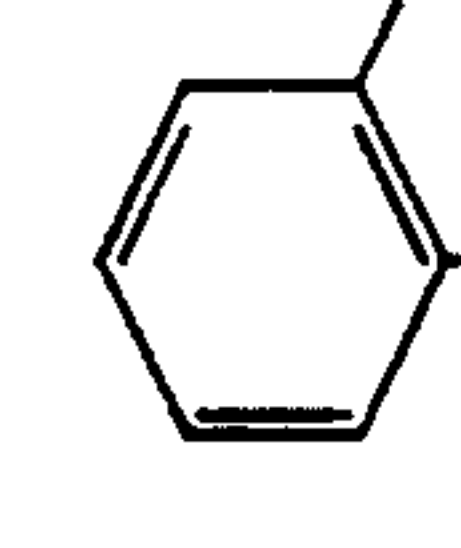
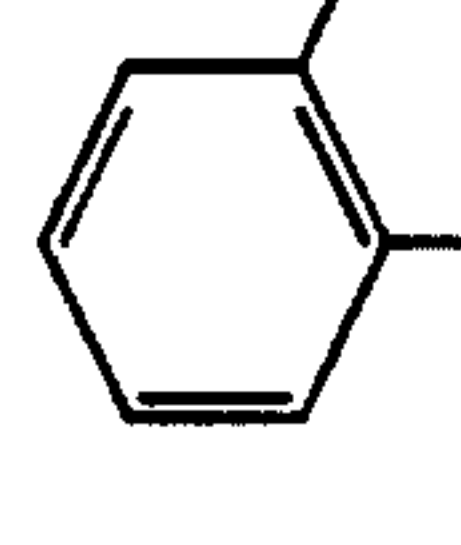
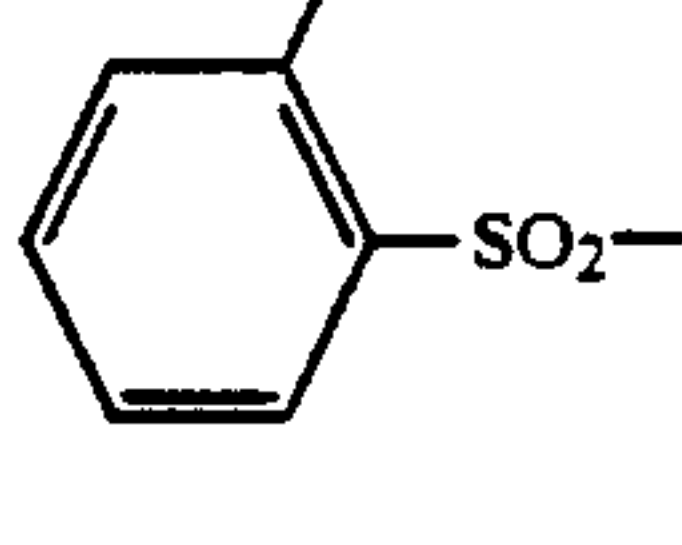
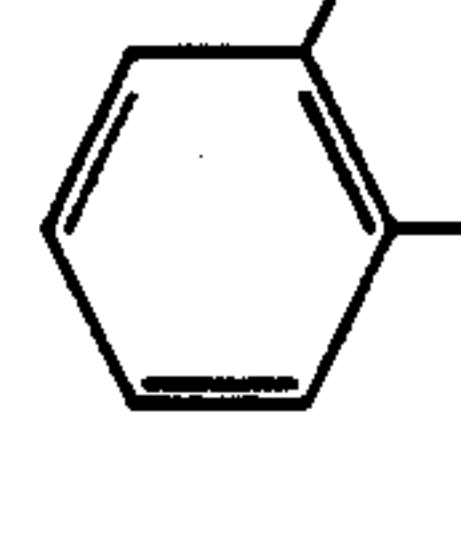
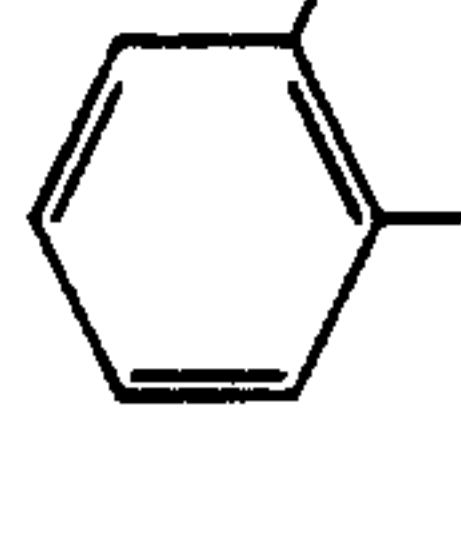
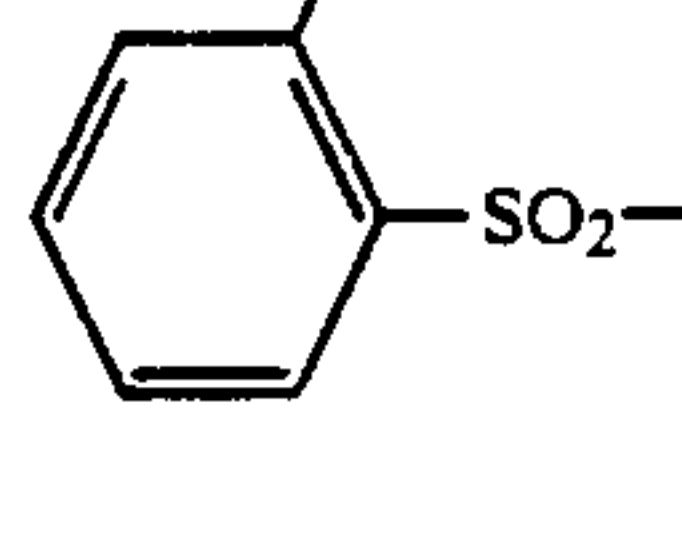
Ex- am- ple No.	Examples of the compounds of the formula (I)							Melting point (°C.)
	R ¹	R ²	R ³	R ⁴	X	Y	Z	
78		H	H	OCH ₃	N	CH	C—Cl	
79			H	OCH ₃	N	CH	C—Cl	
80		H	H	OCH ₃	N	CH	C—Cl	
81		H	H	OCH ₃	N	CH	C—Cl	
82			H	OCH ₃	N	CH	C—Cl	
83			H	OCH ₃	N	CH	C—Cl	
84		H	H	OCH ₃	N	CH	C—Cl	
85			H	OCH ₃	N	CH	C—OCHF ₂	
86		H	H	OCH ₃	N	CH	C—OCHF ₂	
87			H	OCH ₃	N	CH	C—OCHF ₂	

TABLE 2-continued

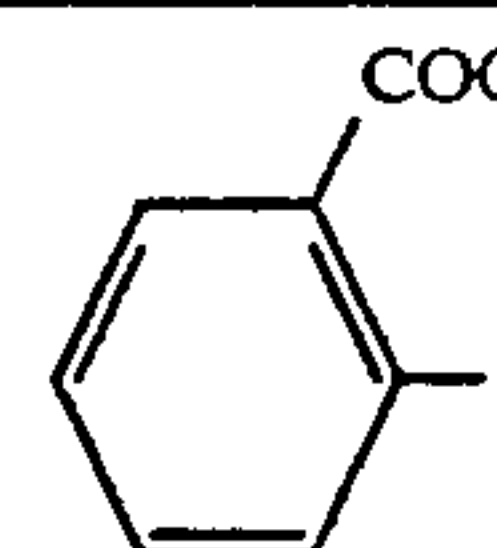
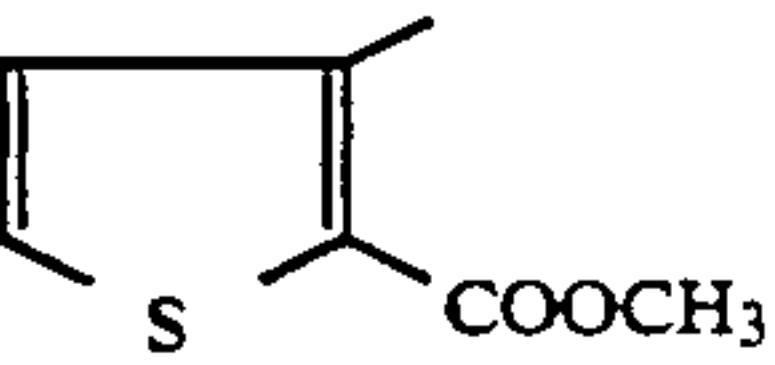
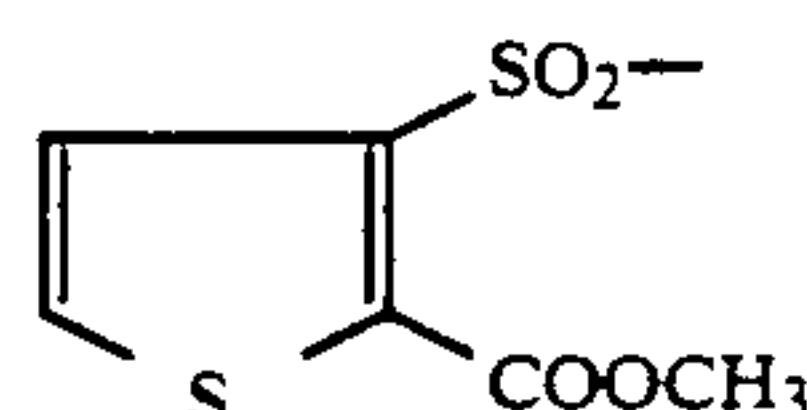
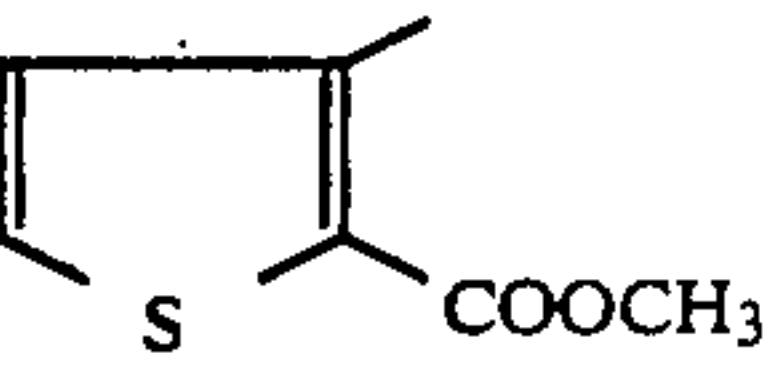
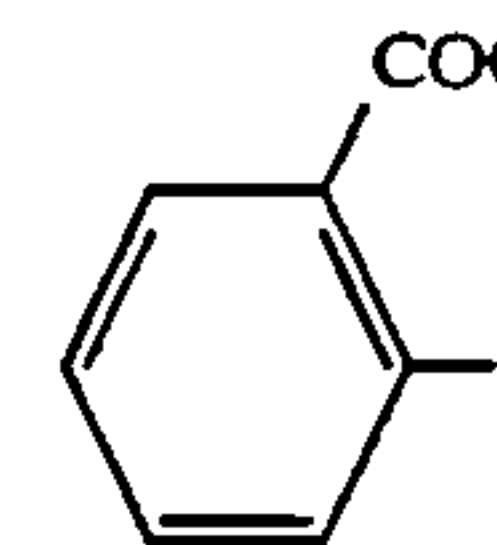
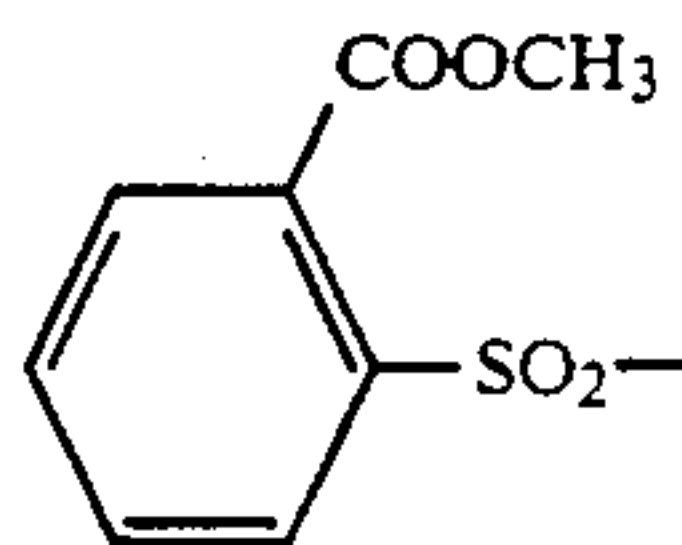
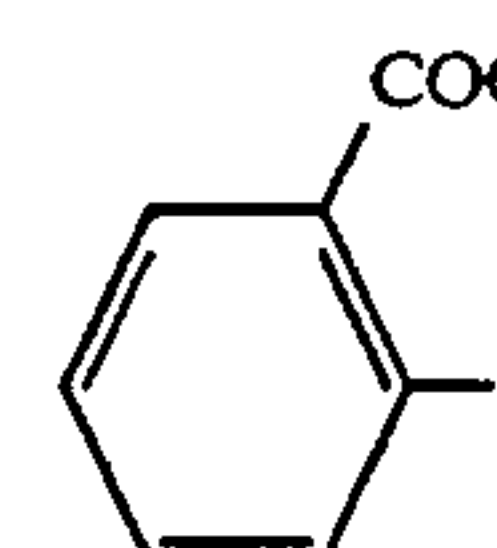
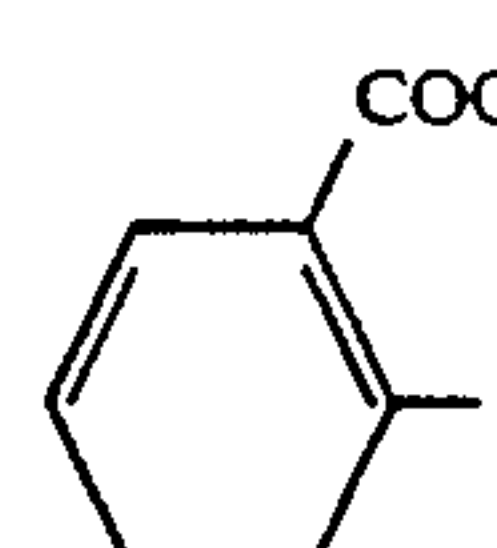
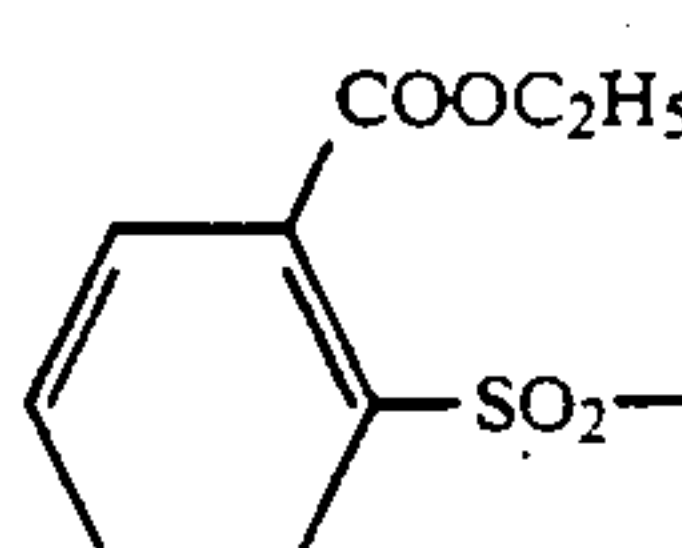
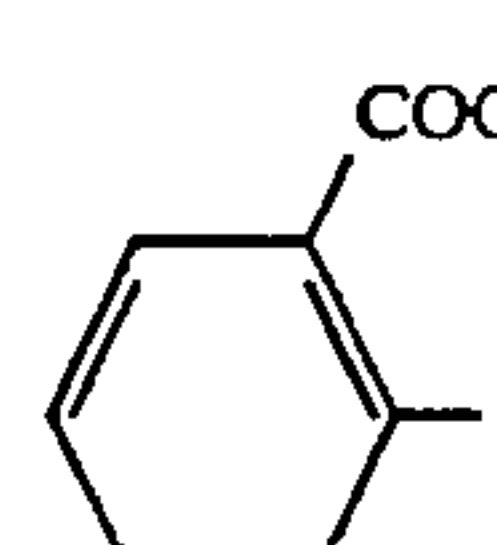
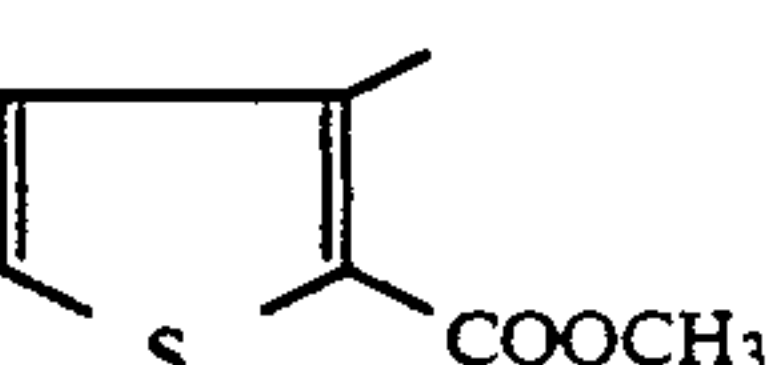
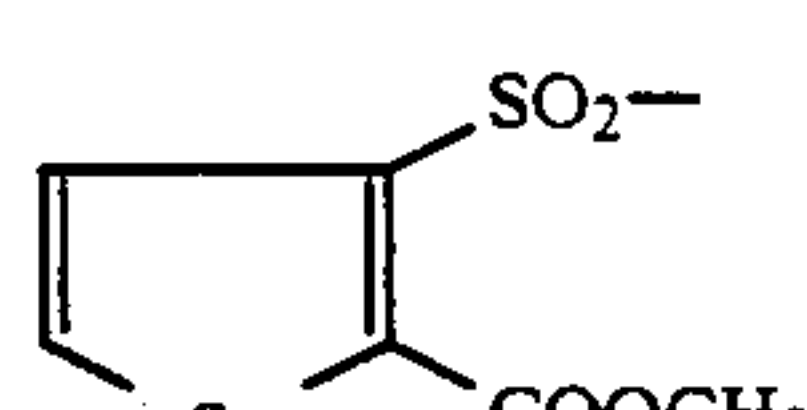
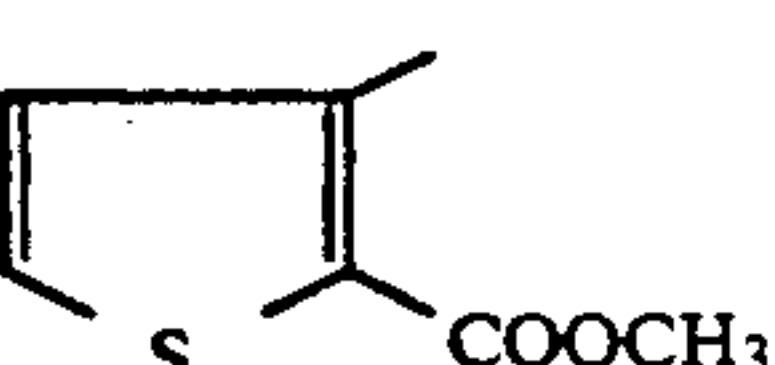
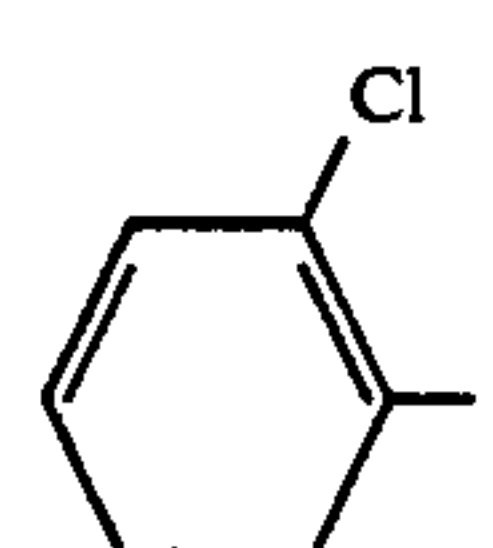
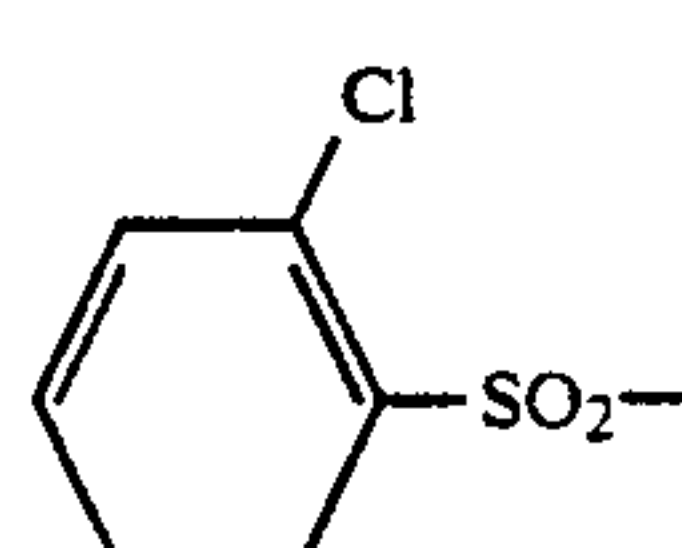
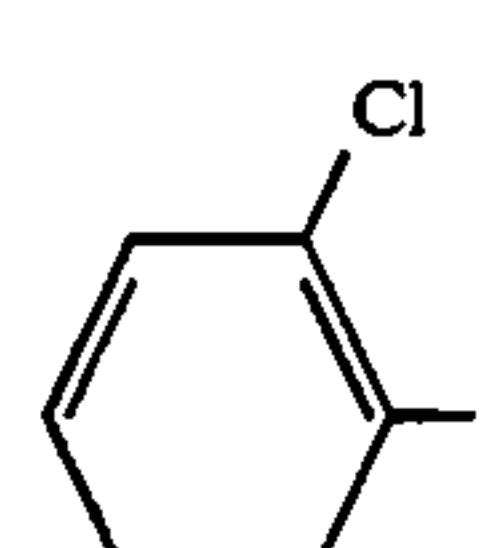
Ex- am- ple No.	Examples of the compounds of the formula (I)							Melting point (°C.)
	R ¹	R ²	R ³	R ⁴	X	Y	Z	
88		H	H	OCH ₃	N	CH	C—OCHF ₂	
89			H	OCH ₃	N	CH	C—OCHF ₂	
90		H	H	OCH ₃	N	CH	C—OCHF ₂	
91			H	OCHF ₂	N	CH	C—OCHF ₂	198
92		H	H	OCHF ₂	N	CH	C—OCHF ₂	180
93			H	OCHF ₂	N	CH	C—OCHF ₂	
94		H	H	OCHF ₂	N	CH	C—OCHF ₂	
95			H	OCHF ₂	N	CH	C—OCHF ₂	
96		H	H	OCHF ₂	N	CH	C—OCHF ₂	
97			H	OCHF ₂	N	CH	C—OCHF ₂	
98		H	H	OCHF ₂	N	CH	C—OCHF ₂	

TABLE 2-continued

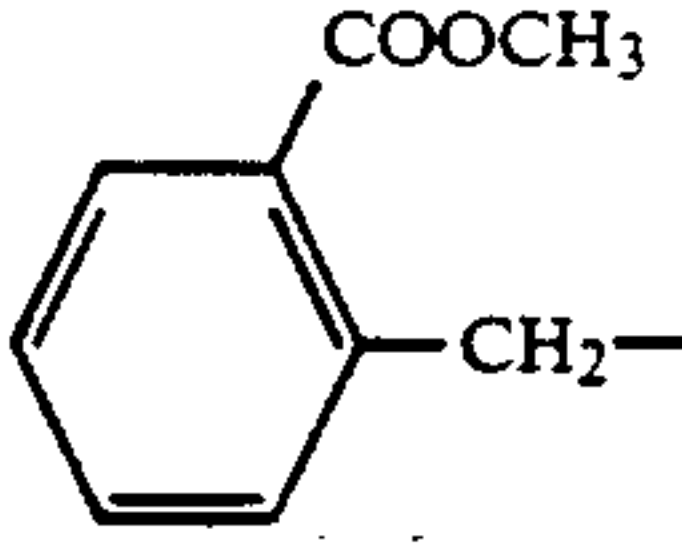
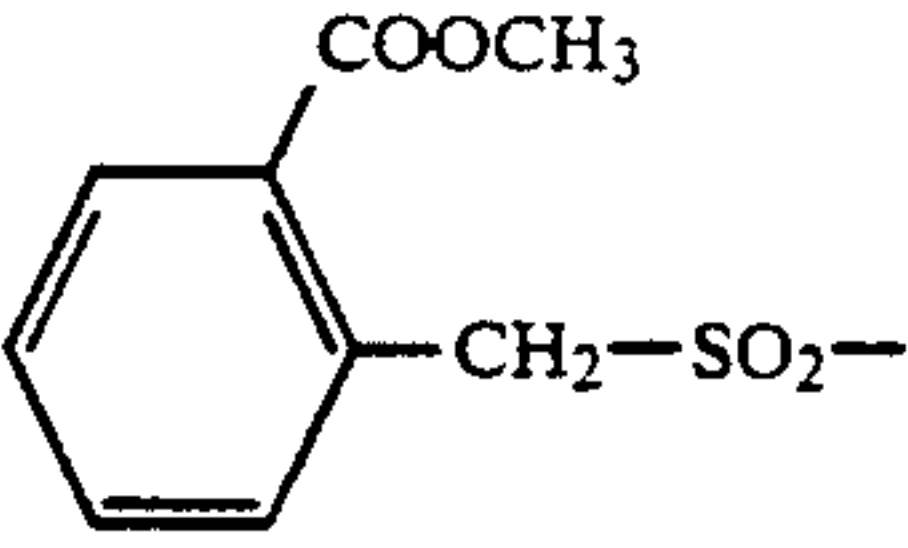
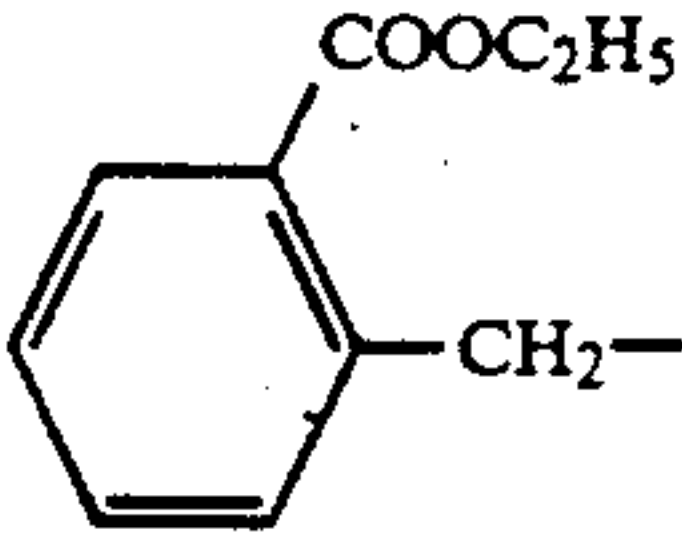
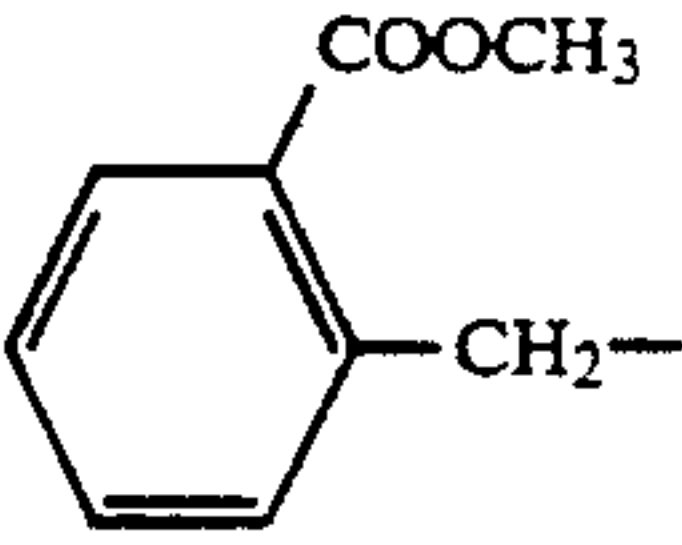
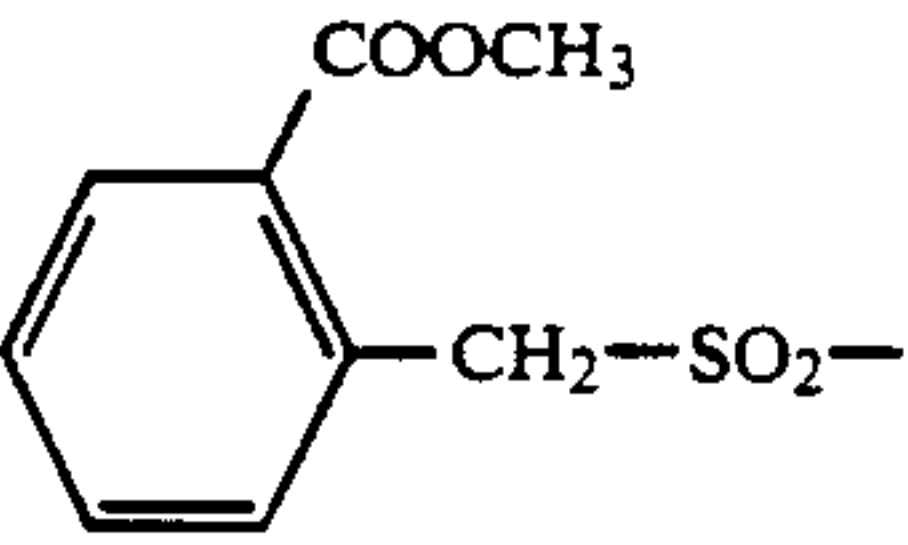
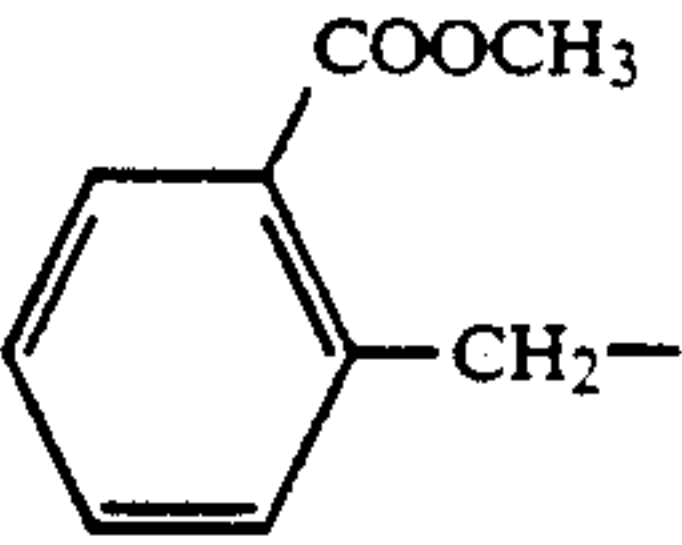
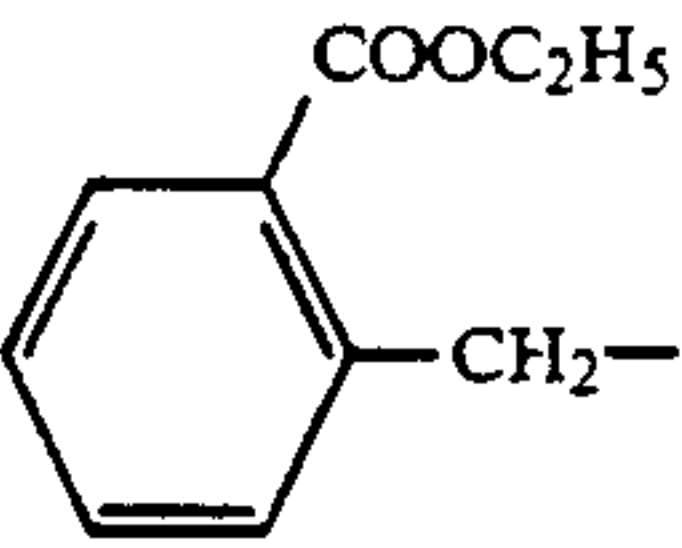
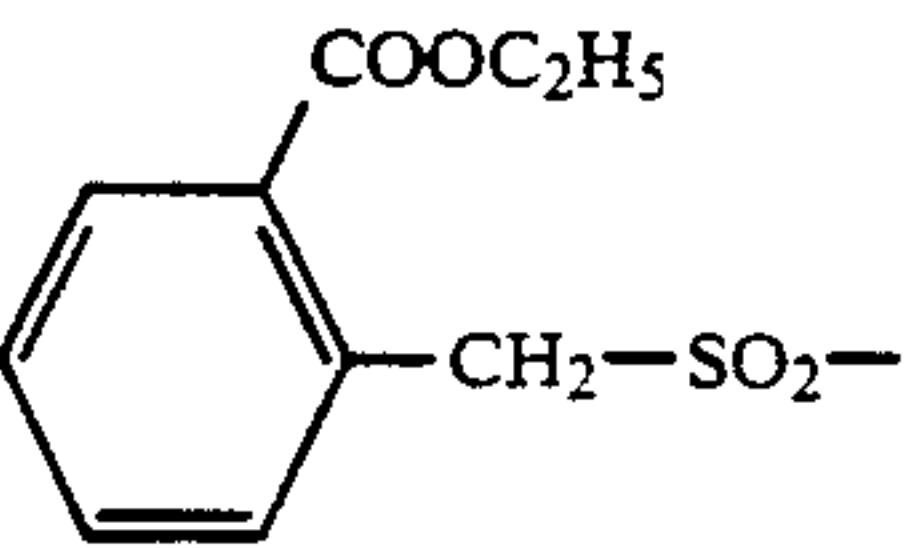
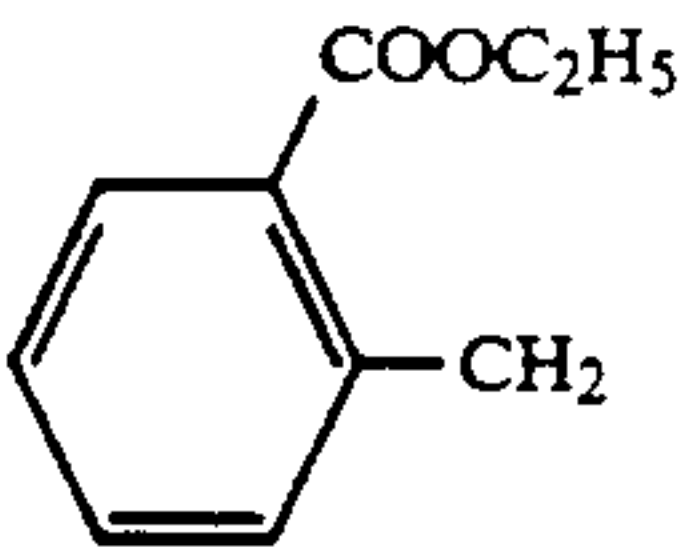
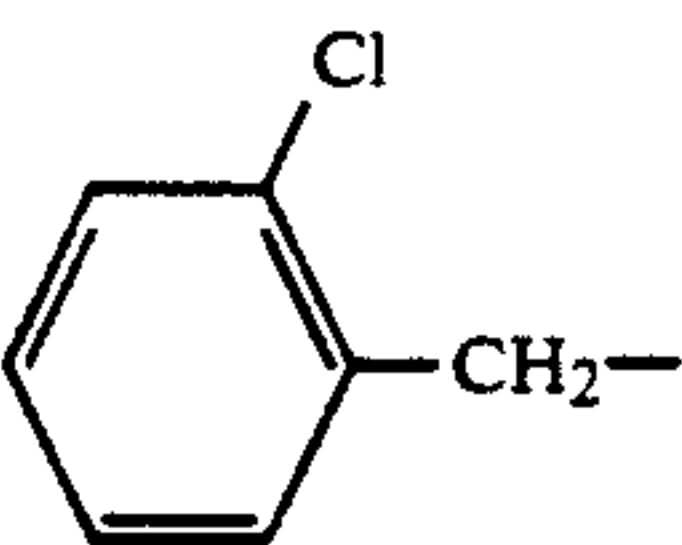
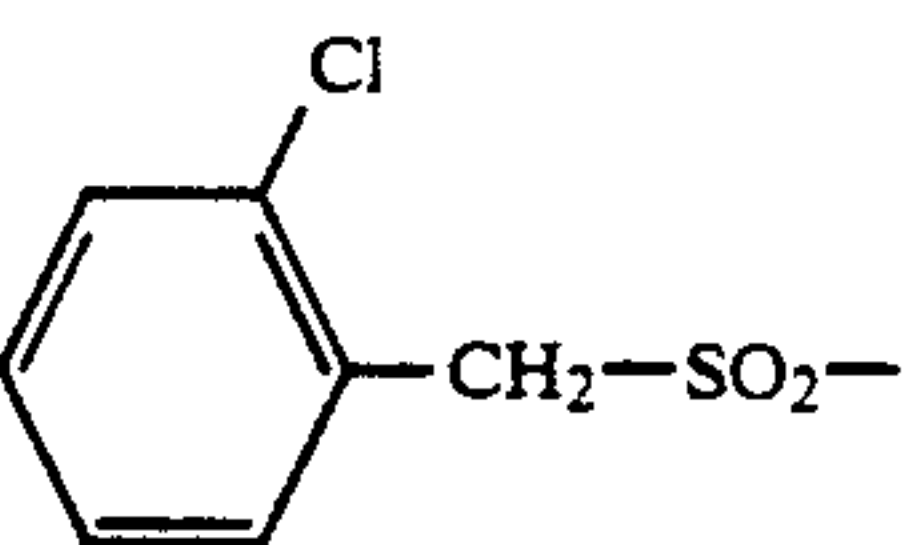
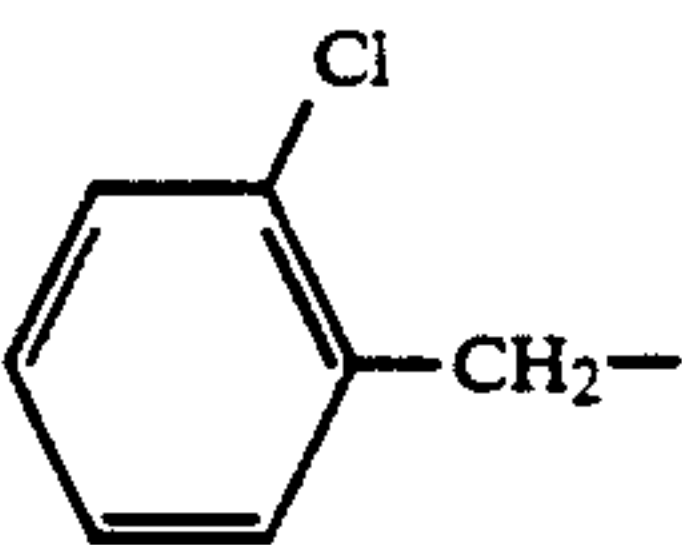
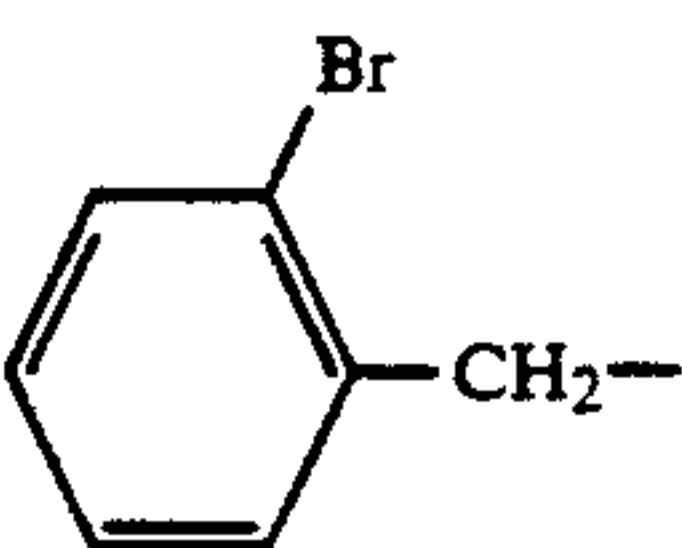
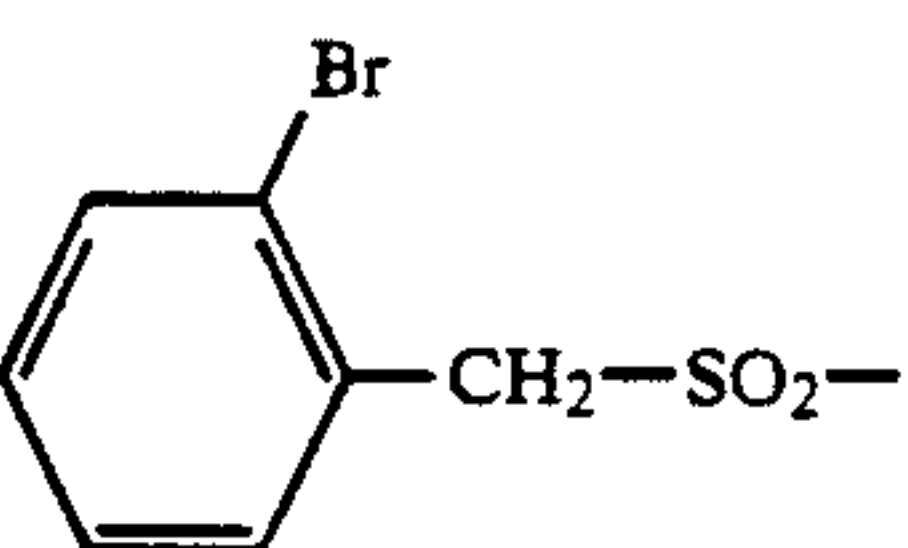
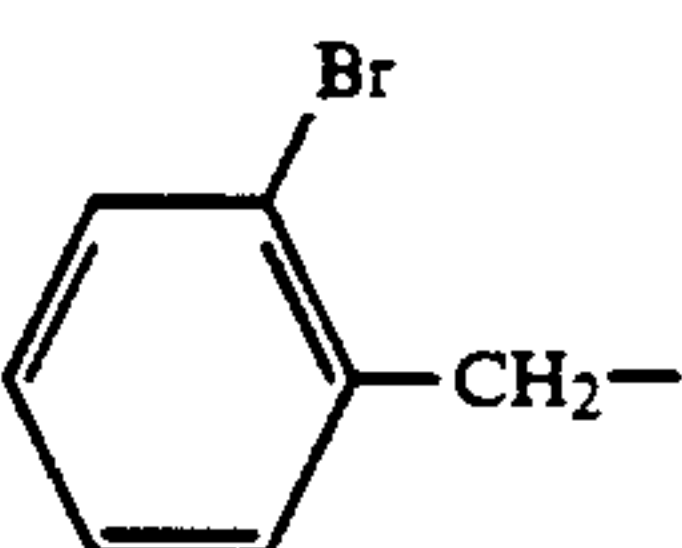
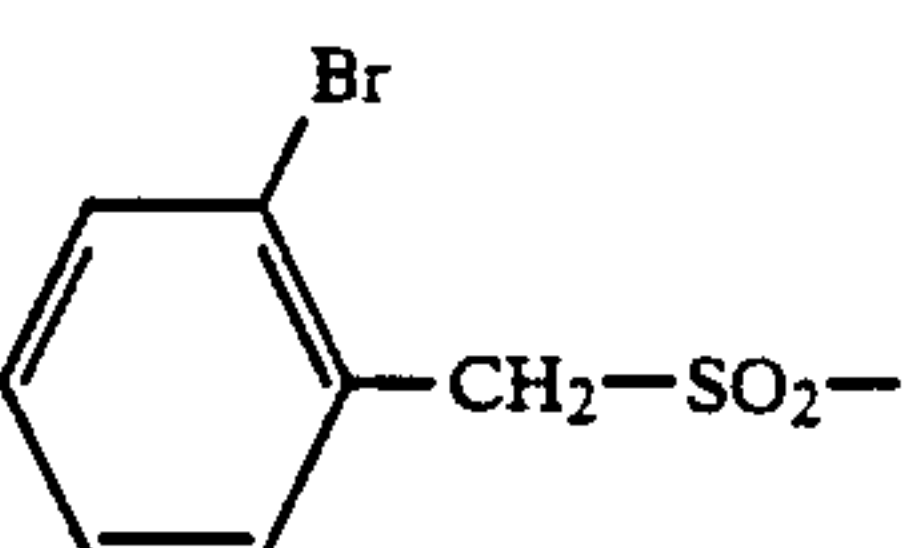
Ex- am- ple No.	Examples of the compounds of the formula (I)							Melting point (°C.)
	R ¹	R ²	R ³	R ⁴	X	Y	Z	
99			H	OCH ₃	N	CH	C—OCH ₃	
100		H	H	OCH ₃	N	CH	C—OCH ₃	
101			H	OCH ₃	N	CH	C—Cl	
102		H	H	OCH ₃	N	CH	C—Cl	
103			H	OCH ₃	N	CH	C—OCH ₃	
104		H	H	OCH ₃	N	CH	C—OCH ₃	
105			H	OCH ₃	N	CH	C—OCH ₃	
106		H	H	OCH ₃	N	CH	C—OCH ₃	
107			H	OCH ₃	N	CH	C—OCH ₃	
108			H	OCH ₃	N	CH	C—OCHF ₂	

TABLE 2-continued

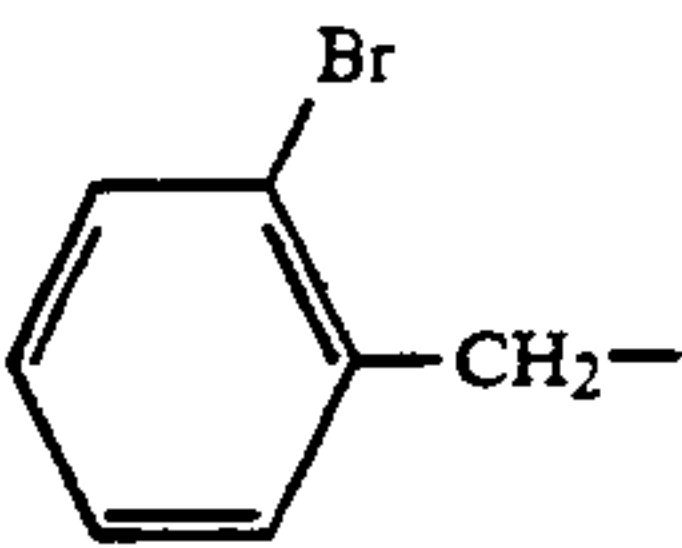
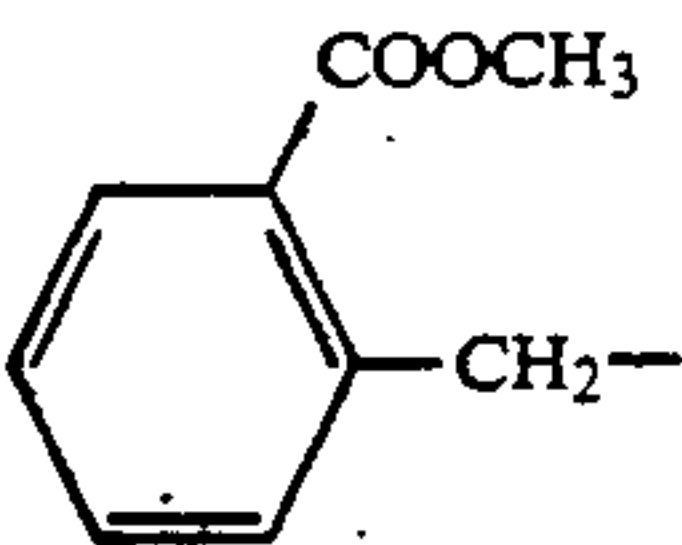
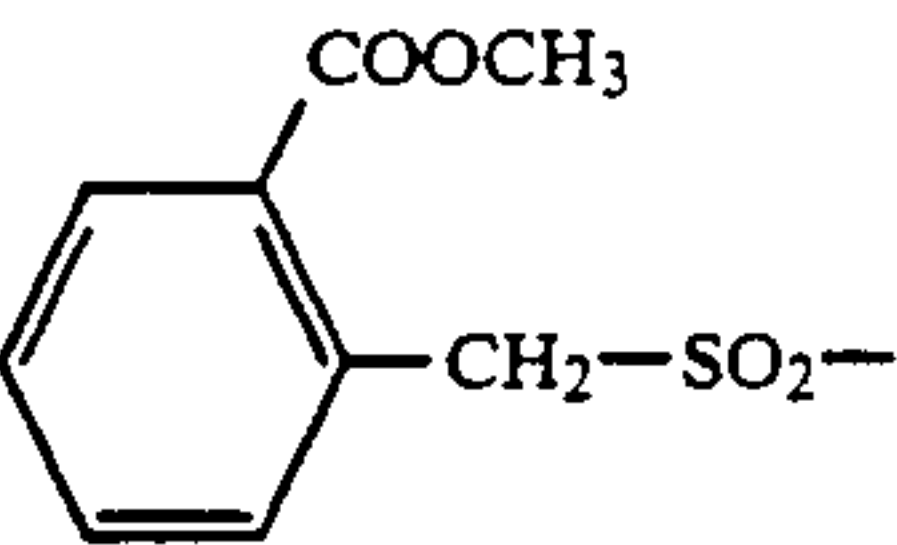
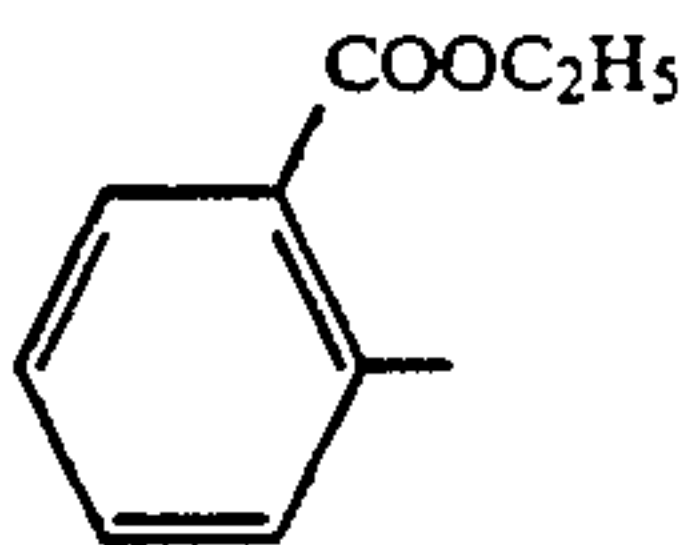
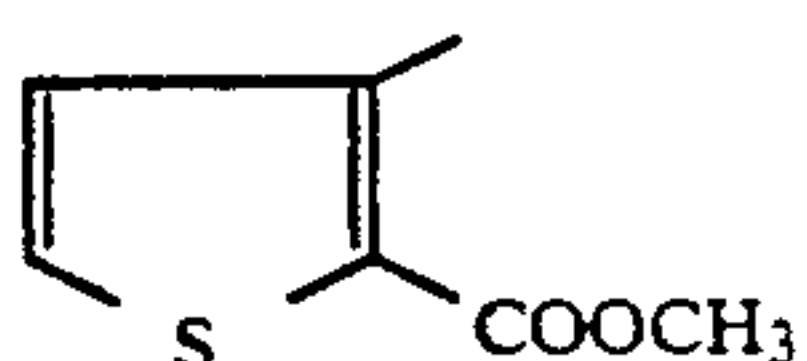
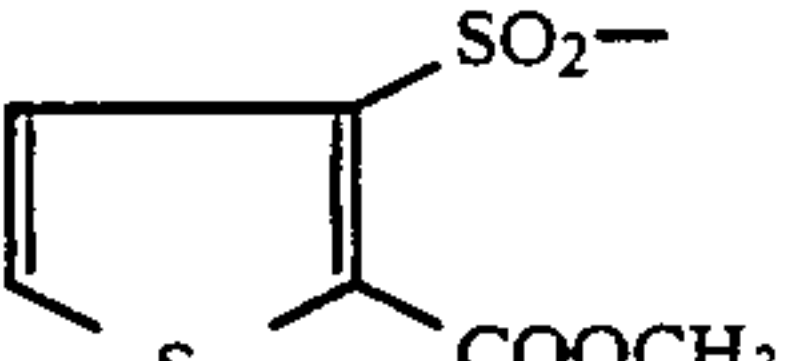
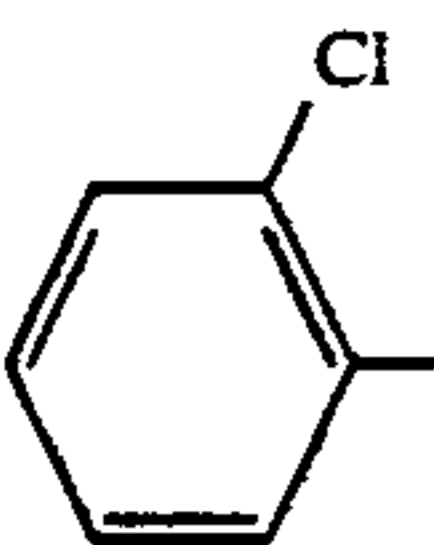
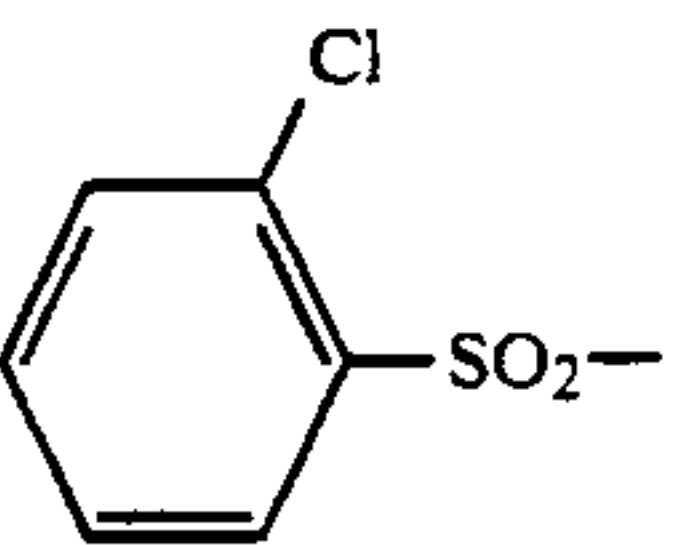
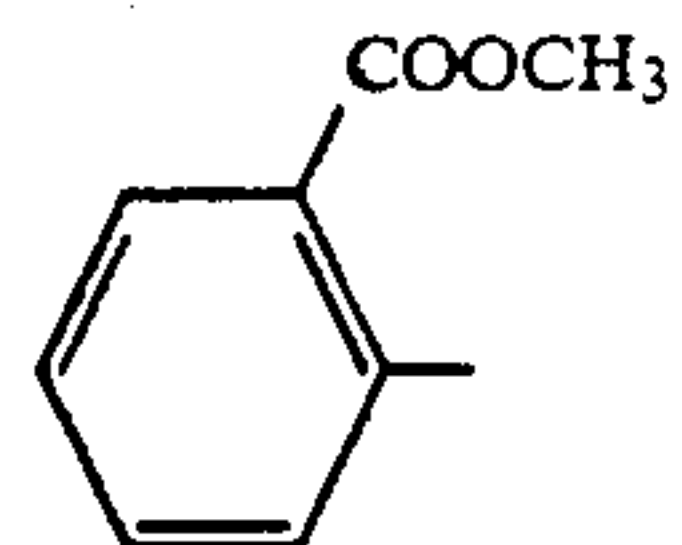
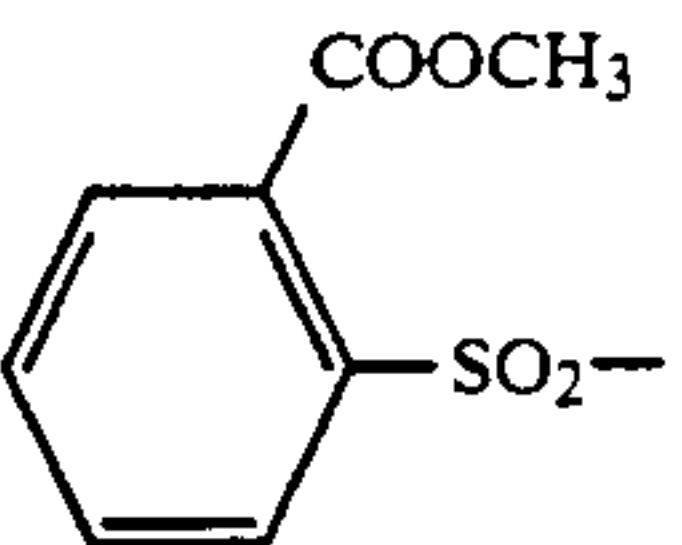
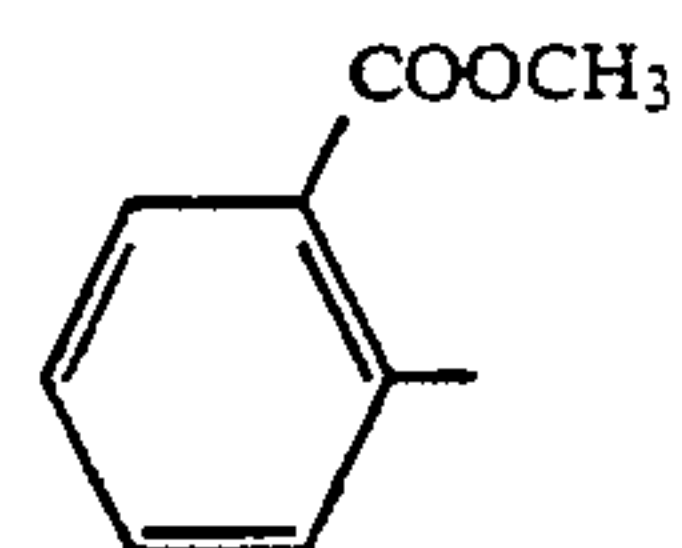
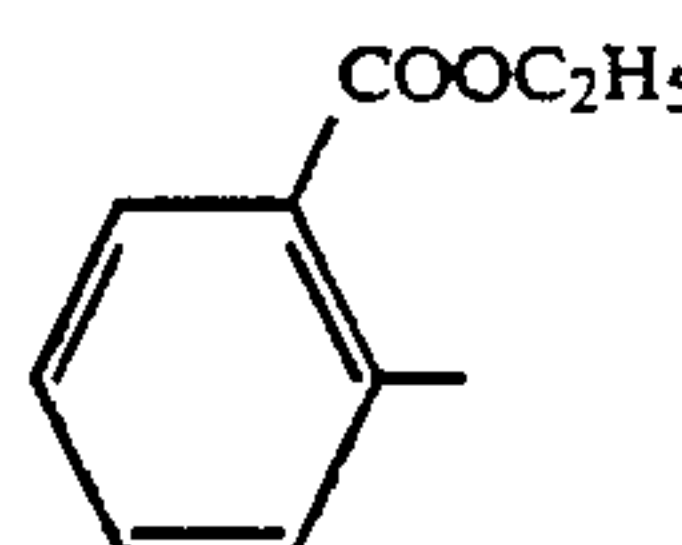
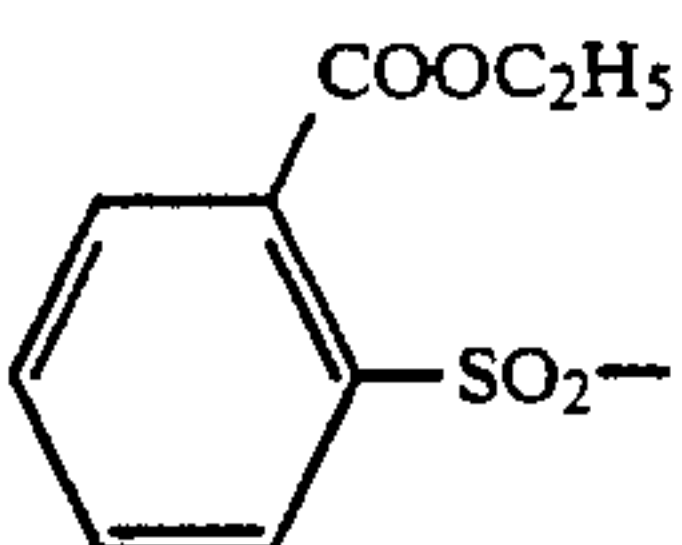
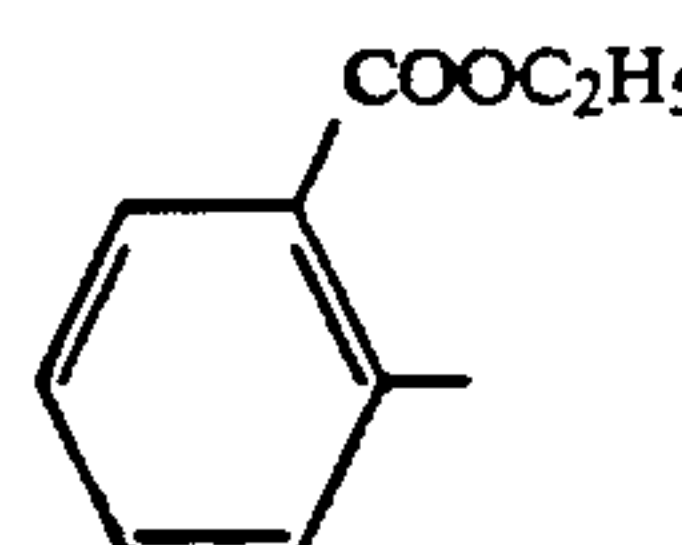
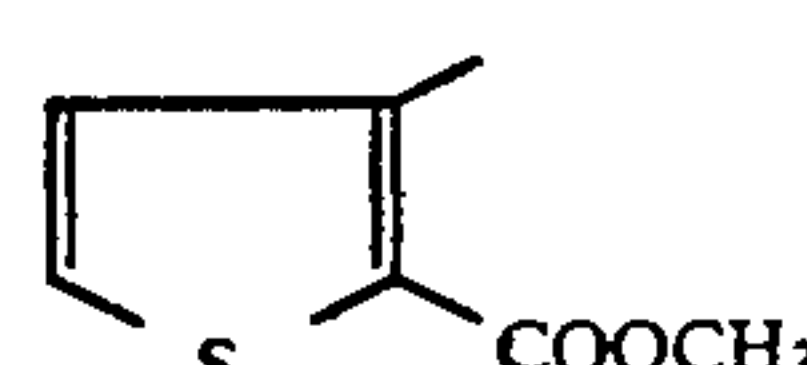
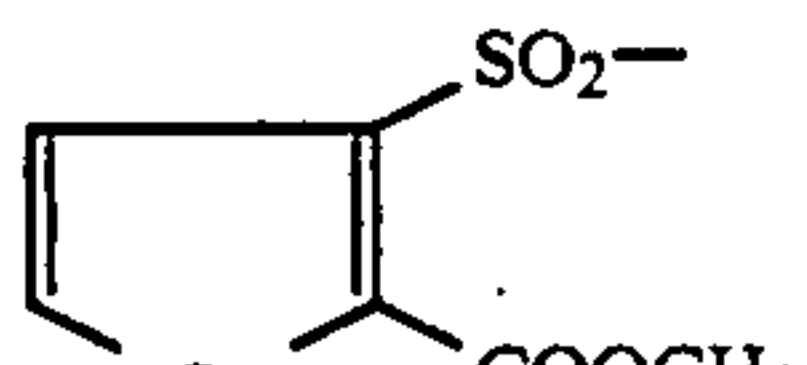
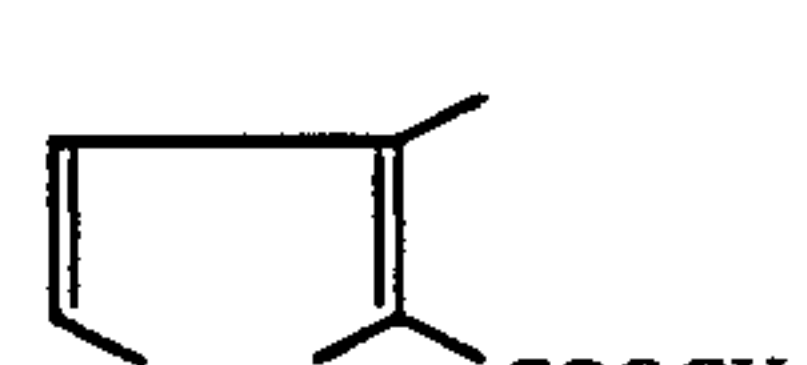
Examples of the compounds of the formula (I)								
Ex- am- ple No.	R ¹	R ²	R ³	R ⁴	X	Y	Z	Melting point (°C.)
109		H	H	OCH ₃	N	CH	C—OCH ₃	
110			H	OCH ₃	N	CH	C—OCHF ₂	
111		H	H	CH ₃	N	N	C—CH ₃	
112			H	CH ₃	N	N	C—CH ₃	
113			H	CH ₃	N	N	C—CH ₃	
114			H	CH ₃	N	N	C—OCH ₃	
115		H	H	CH ₃	N	N	C—OCH ₃	
116			H	CH ₃	N	N	C—OCH ₃	
117		H	H	CH ₃	N	N	C—OCH ₃	
118			H	CH ₃	N	N	C—OCH ₃	
119		H	H	CH ₃	N	N	C—OCH ₃	

TABLE 2-continued

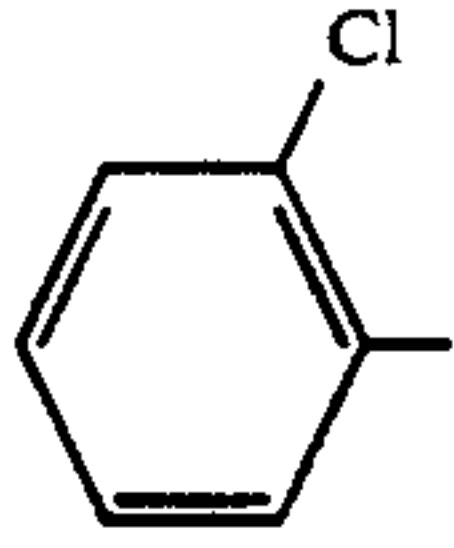
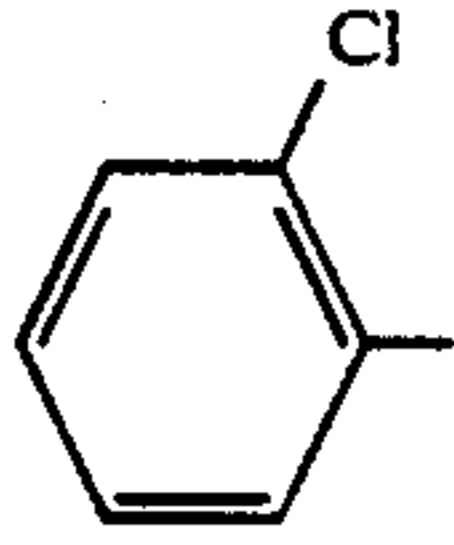
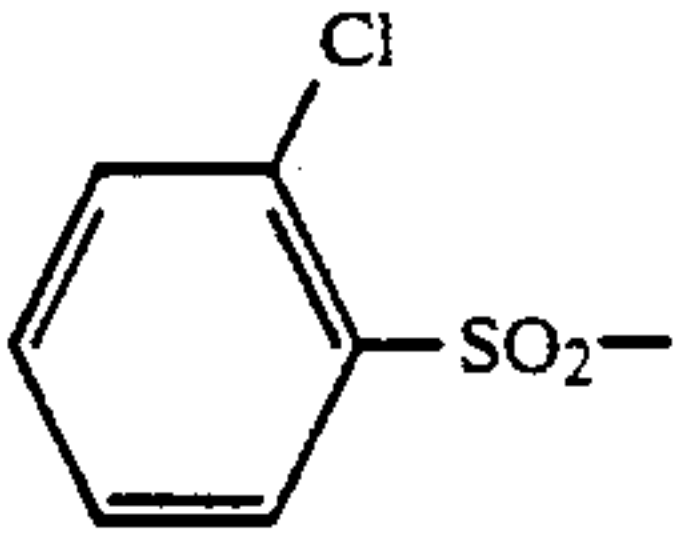
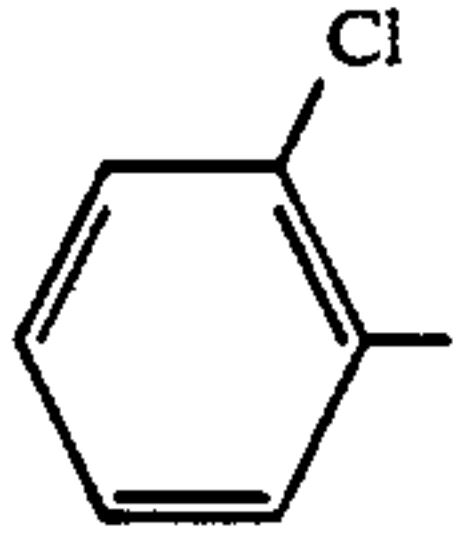
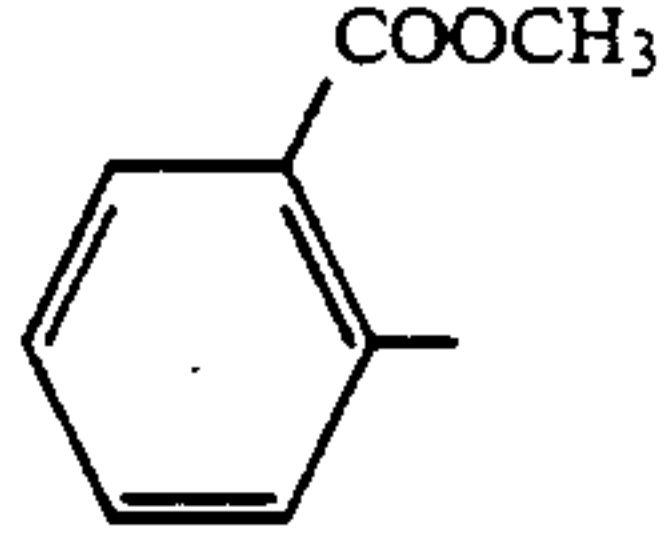
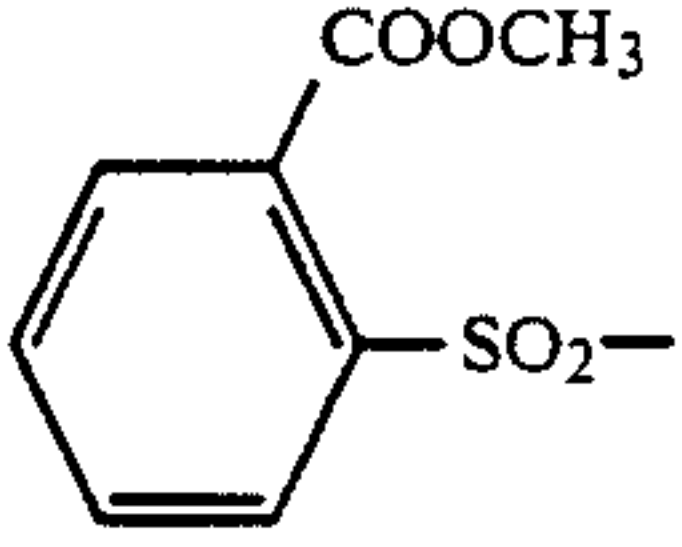
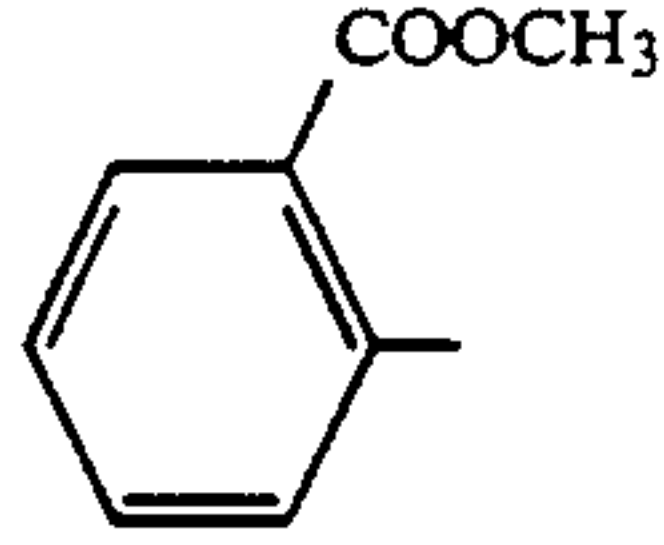
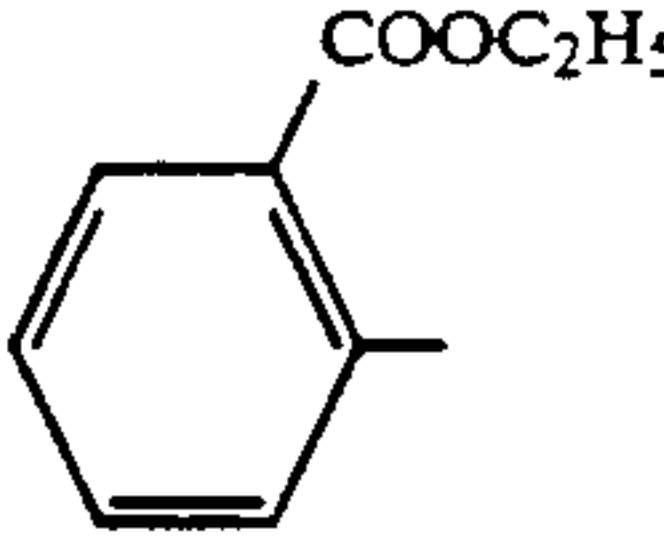
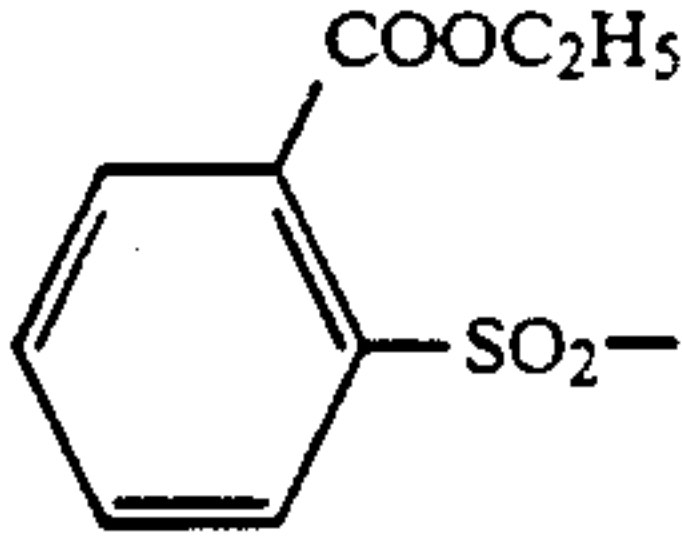
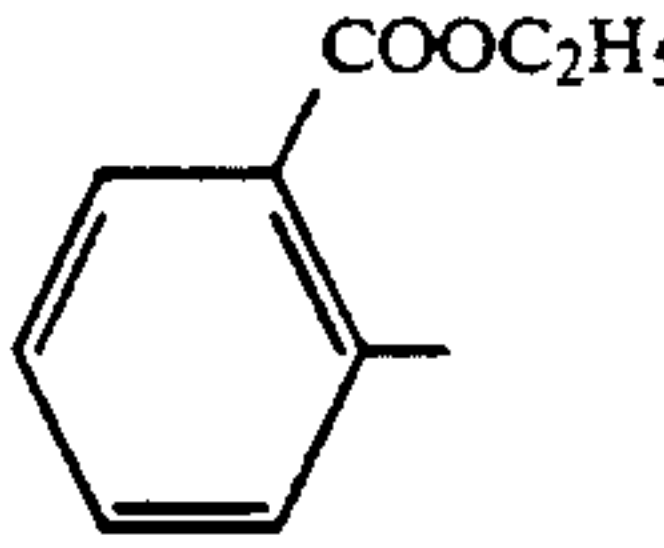
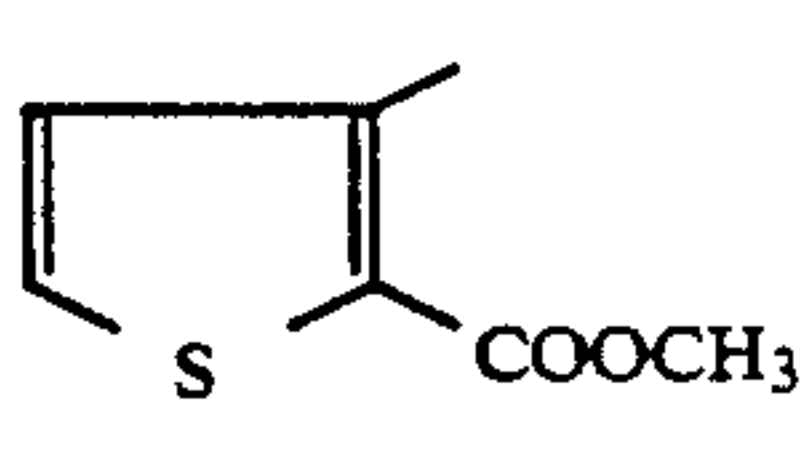
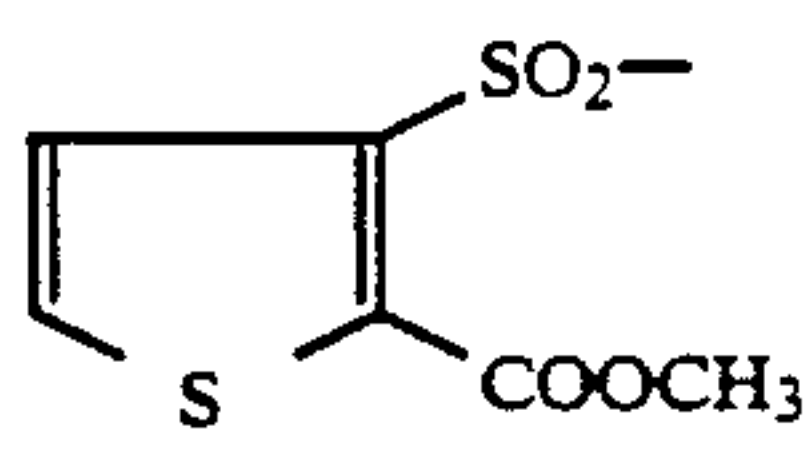
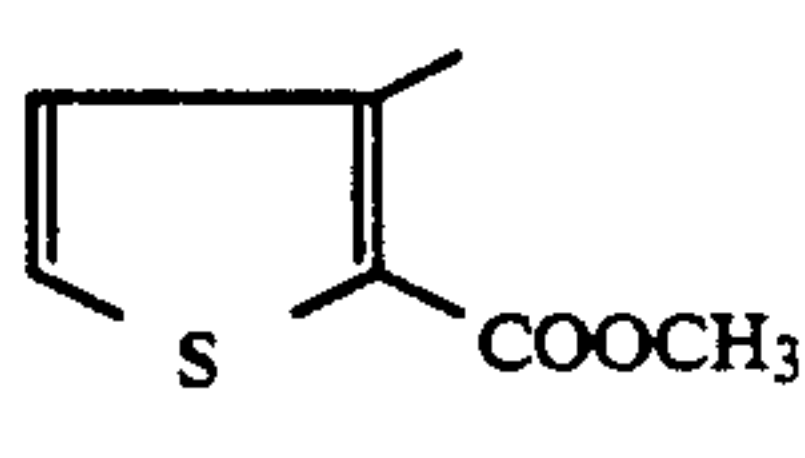
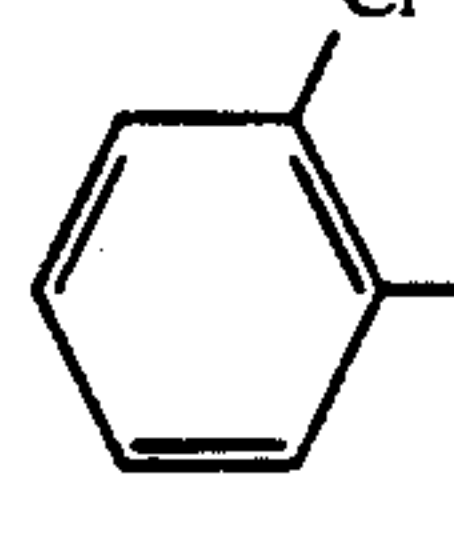
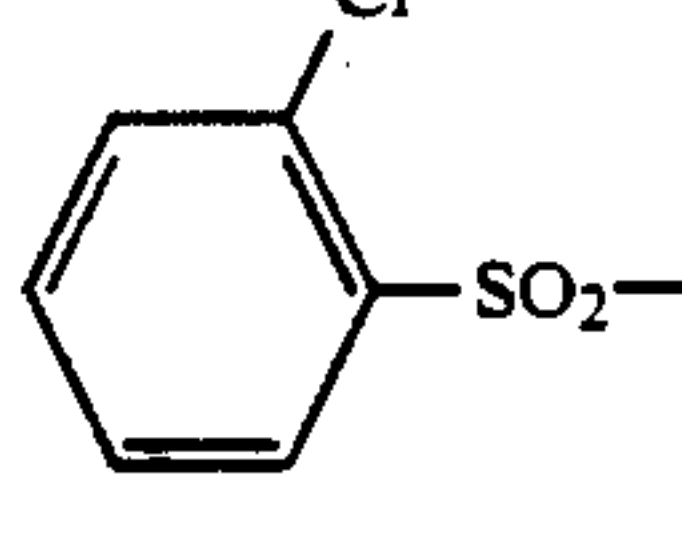
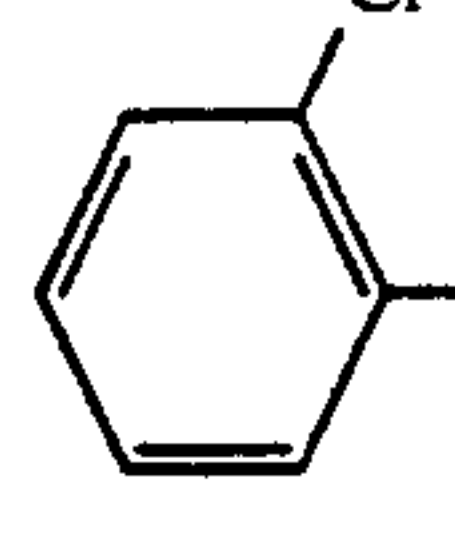
Ex- am- ple No.	Examples of the compounds of the formula (I)							Melting point (°C.)
	R ¹	R ²	R ³	R ⁴	X	Y	Z	
120		Na	H	CH ₃	N	N	C—OCH ₃	
121			H	CH ₃	N	N	C—OCH ₃	
122		H	H	CH ₃	N	N	C—OCH ₃	
123			H	OCH ₃	N	N	C—OCH ₃	
124		H	H	OCH ₃	N	N	C—OCH ₃	
125			H	OCH ₃	N	N	C—OCH ₃	
126		H	H	OCH ₃	N	N	C—OCH ₃	
127			H	OCH ₃	N	N	C—OCH ₃	
128		H	H	OCH ₃	N	N	C—OCH ₃	
129			H	OCH ₃	N	N	C—OCH ₃	
130		H	H	OCH ₃	N	N	C—OCH ₃	

TABLE 2-continued

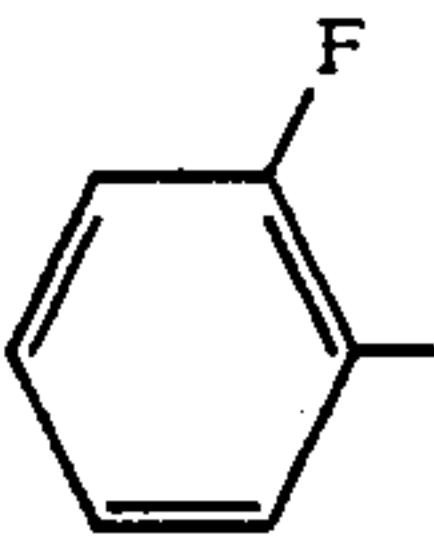
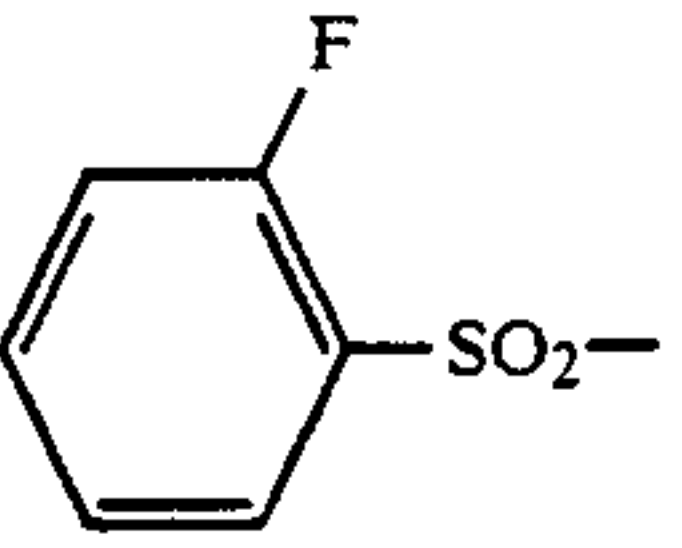
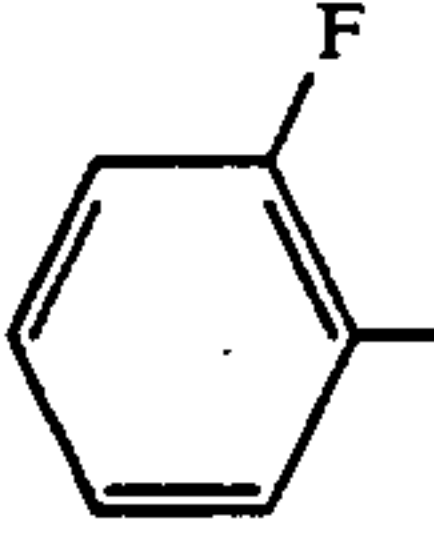
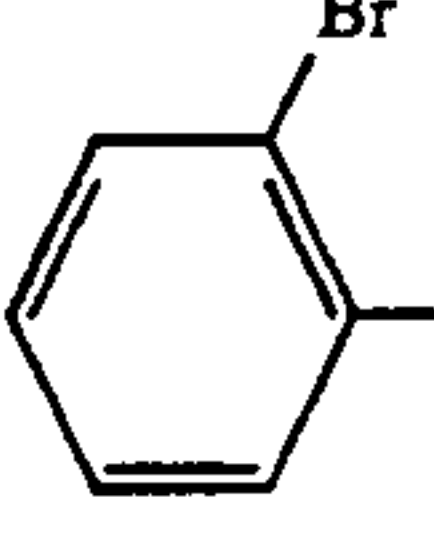
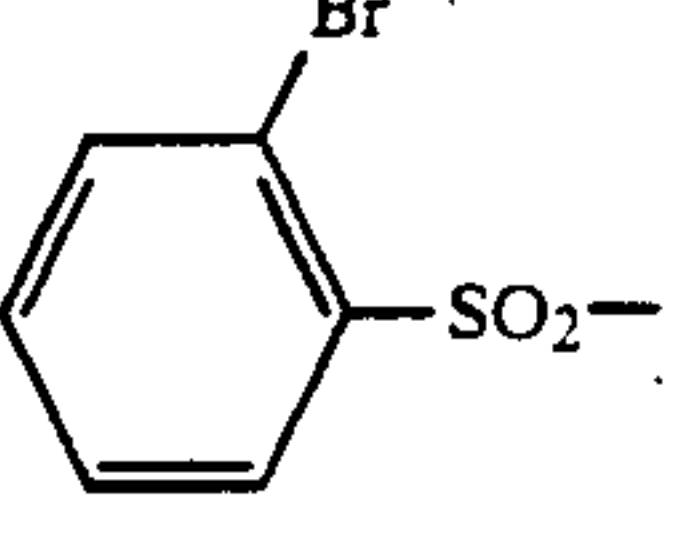
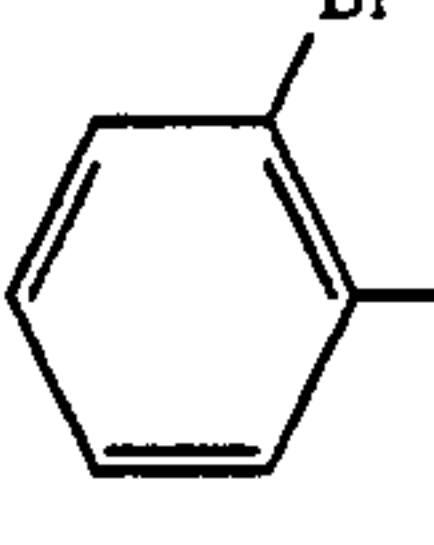
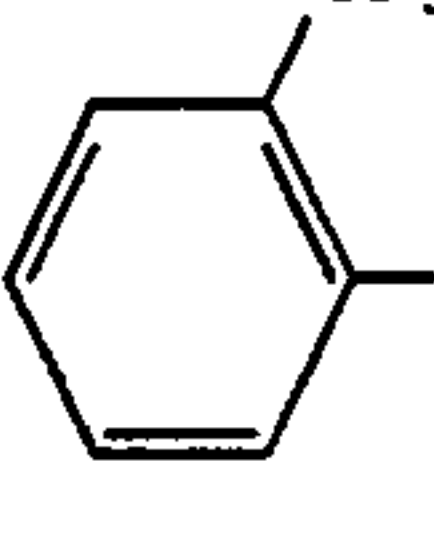
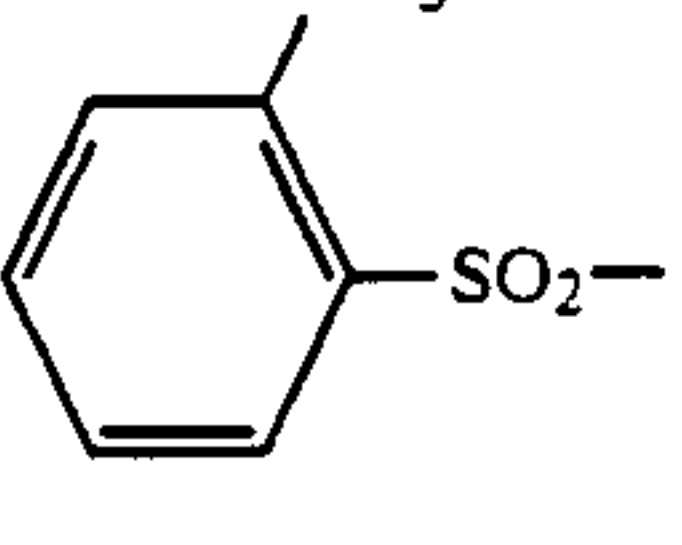
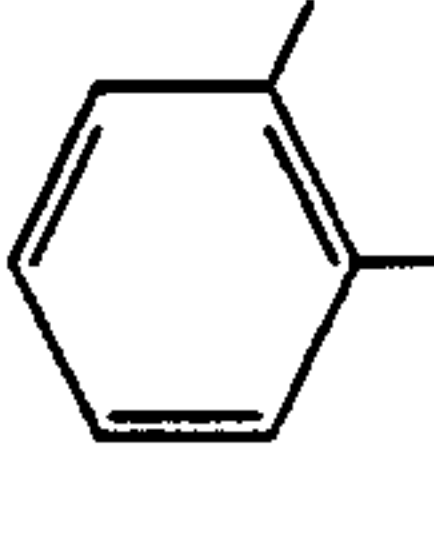
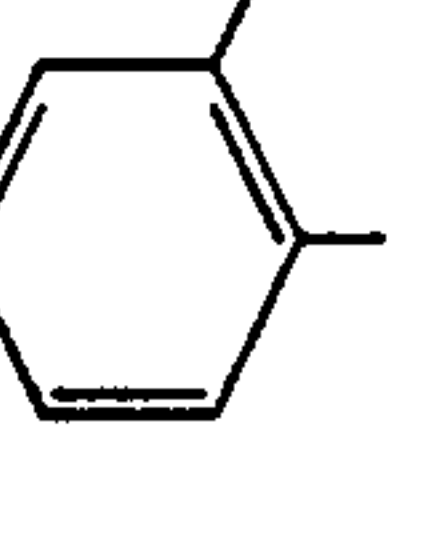
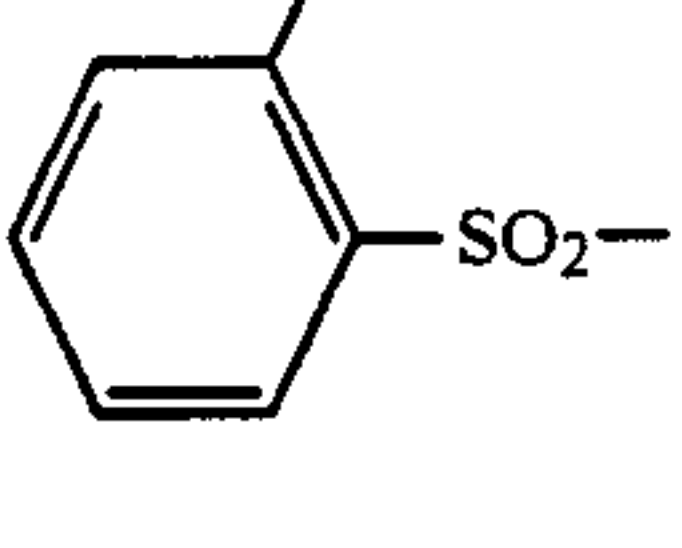
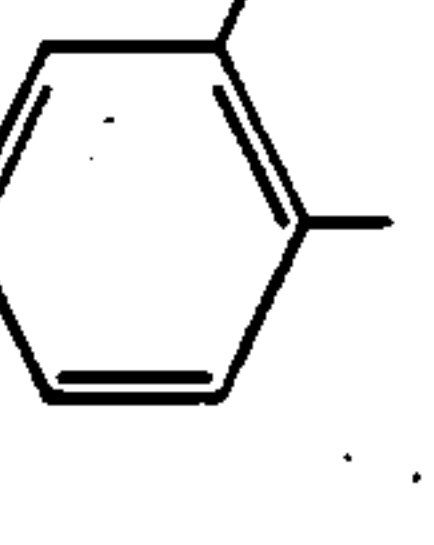
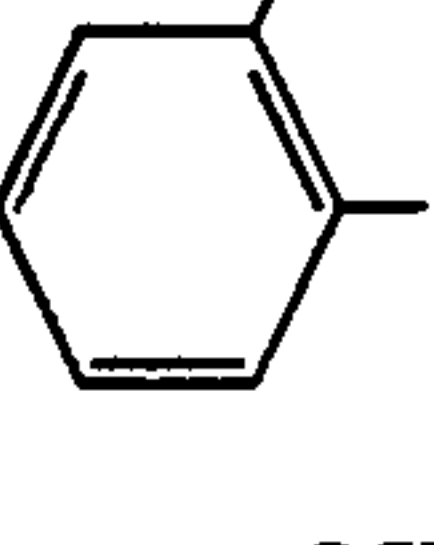
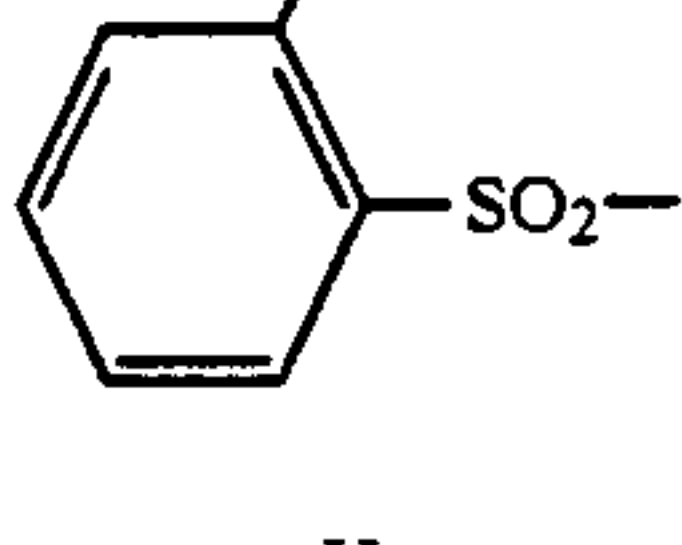
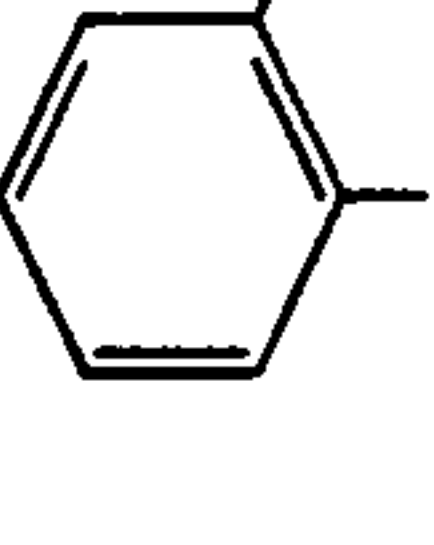
Ex- am- ple No.	Examples of the compounds of the formula (I)							Melting point (°C.)
	R ¹	R ²	R ³	R ⁴	X	Y	Z	
131			H	OCH ₃	N	N	C—OCH ₃	
132		H	H	OCH ₃	N	N	C—OCH ₃	
133			H	OCH ₃	N	N	C—OCH ₃	
134		H	H	OCH ₃	N	N	C—OCH ₃	
135			H	OCH ₃	N	N	C—OCH ₃	
136		H	H	OCH ₃	N	N	C—OCH ₃	
137			H	OCH ₃	N	N	C—OCH ₃	
138		H	H	OCH ₃	N	N	C—OCH ₃	
139			H	OCH ₃	N	N	C—OCH ₃	
140		H	H	OCH ₃	N	N	C—OCH ₃	201

TABLE 2-continued

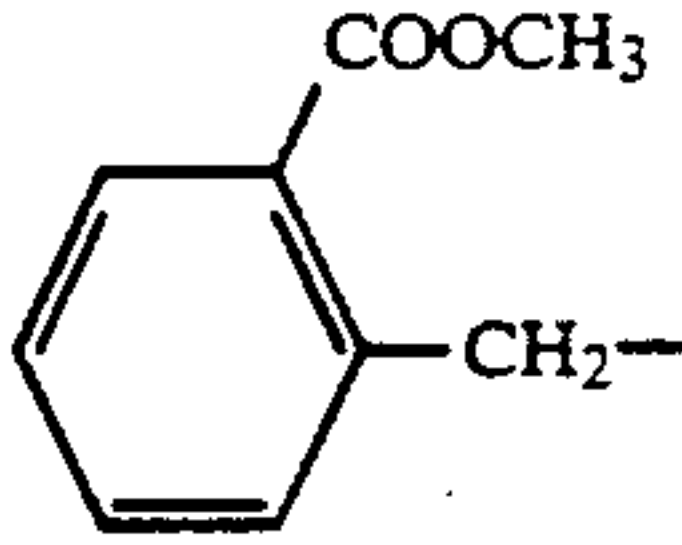
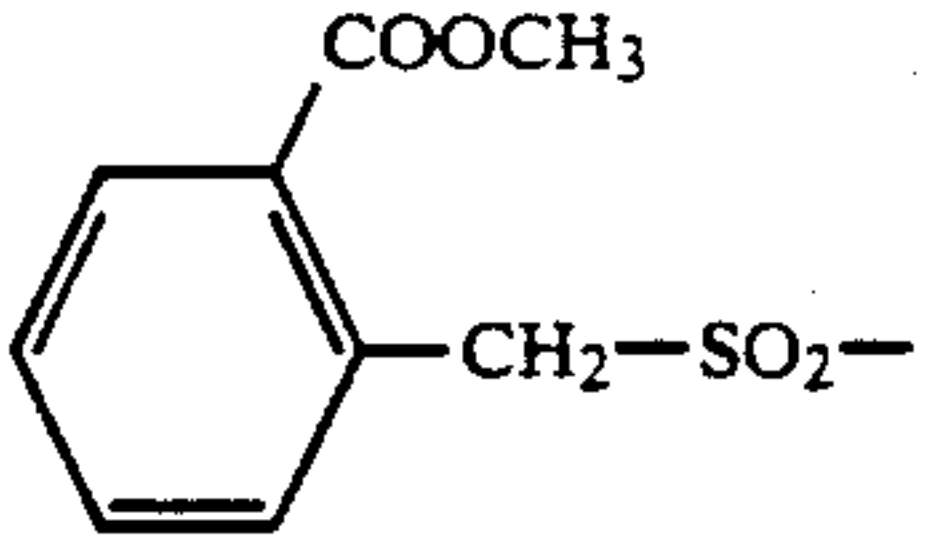
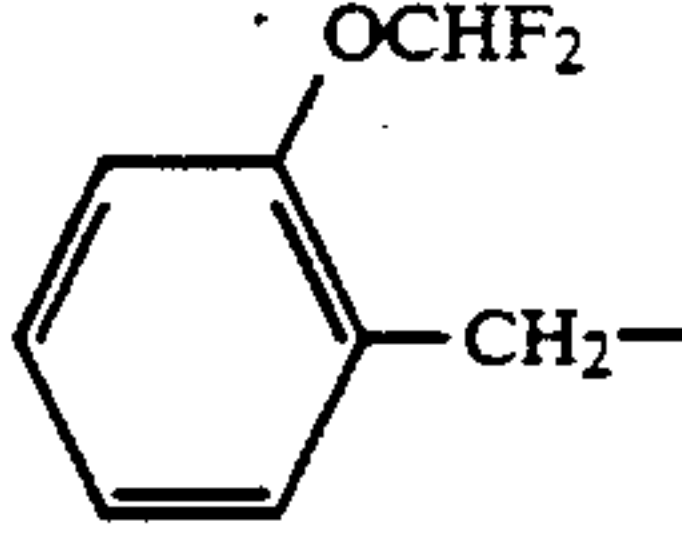
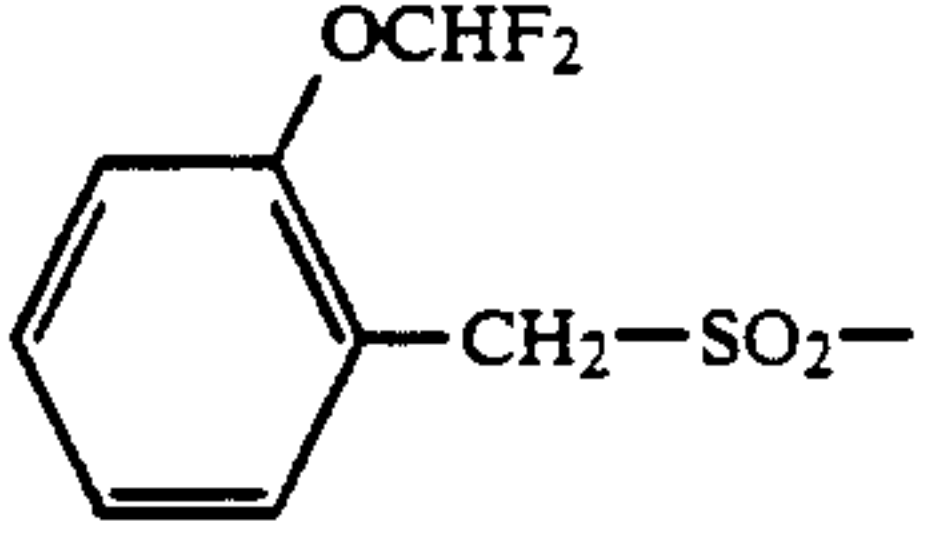
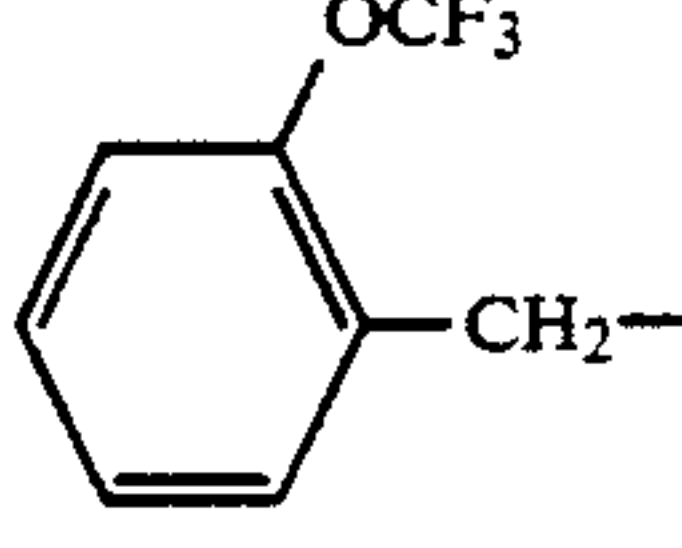
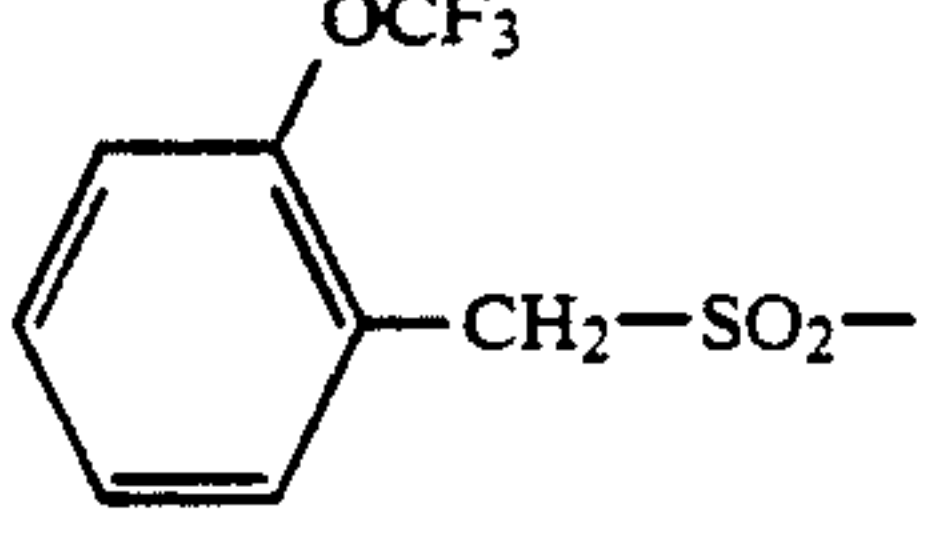
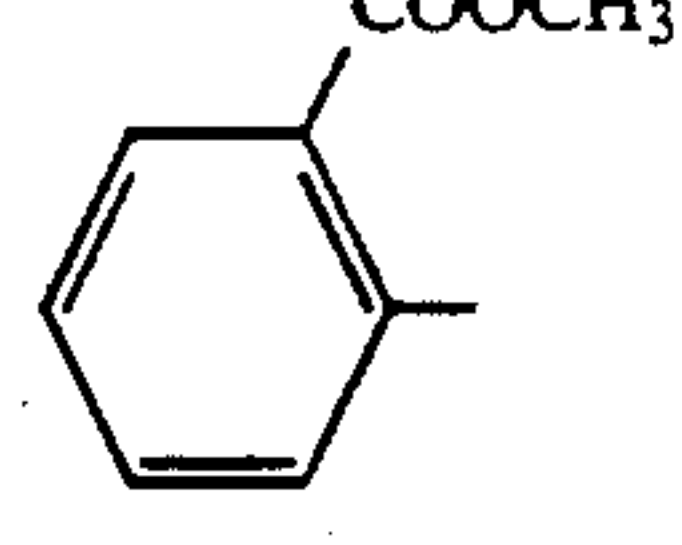
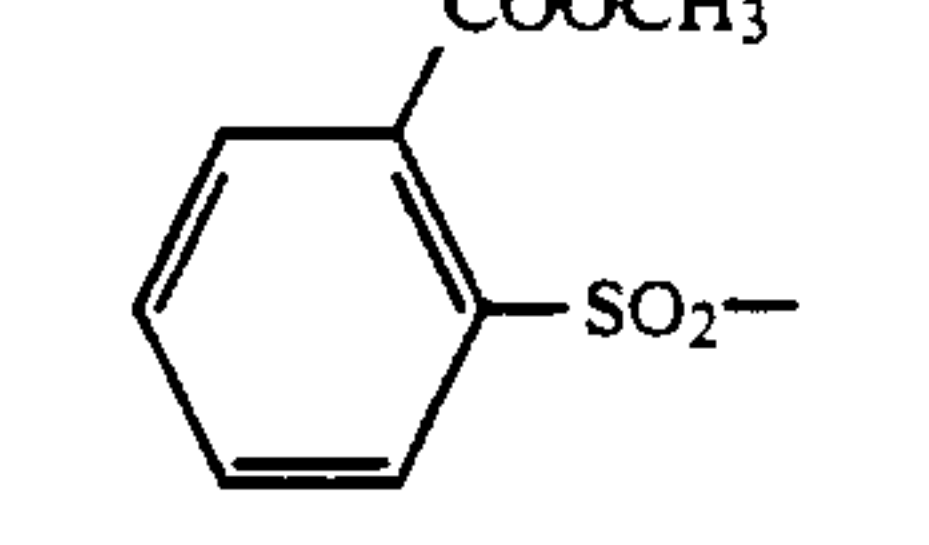
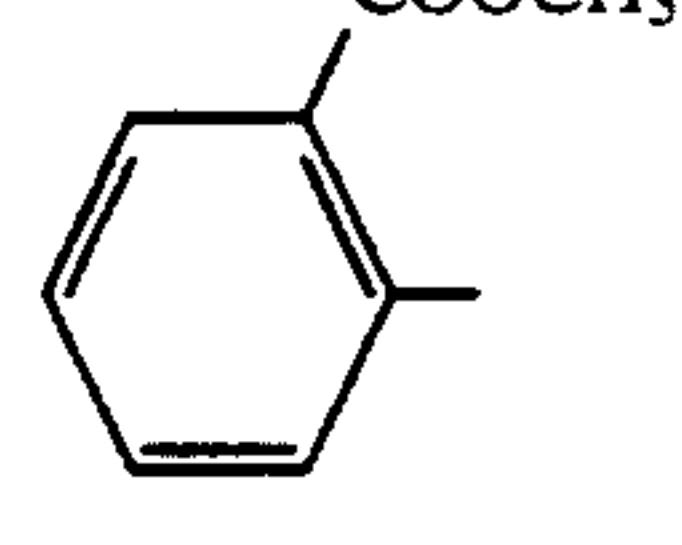
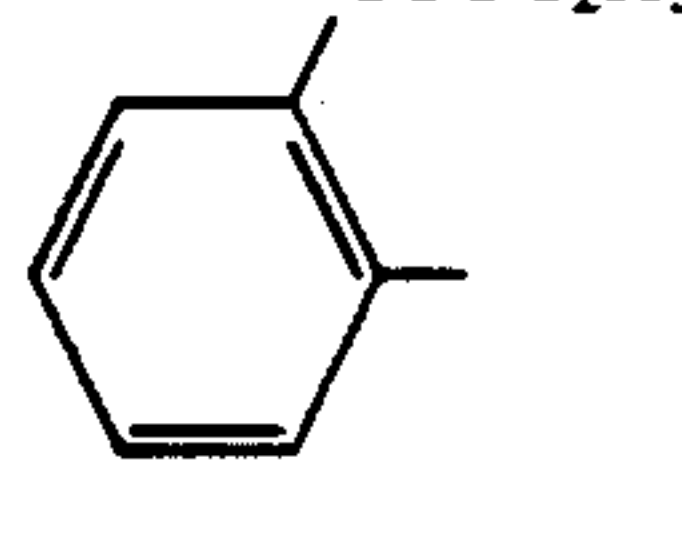
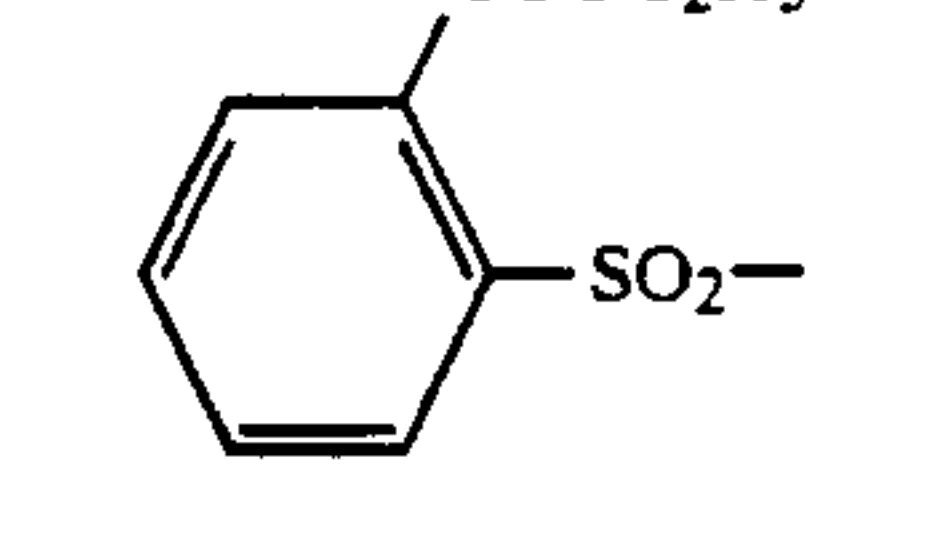
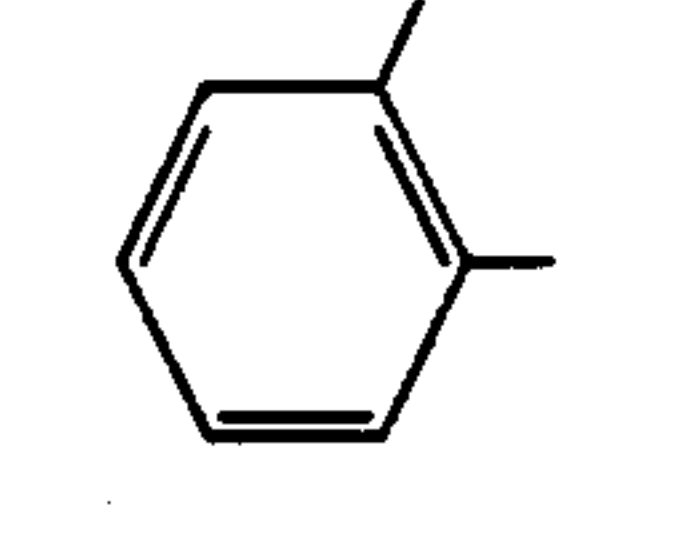
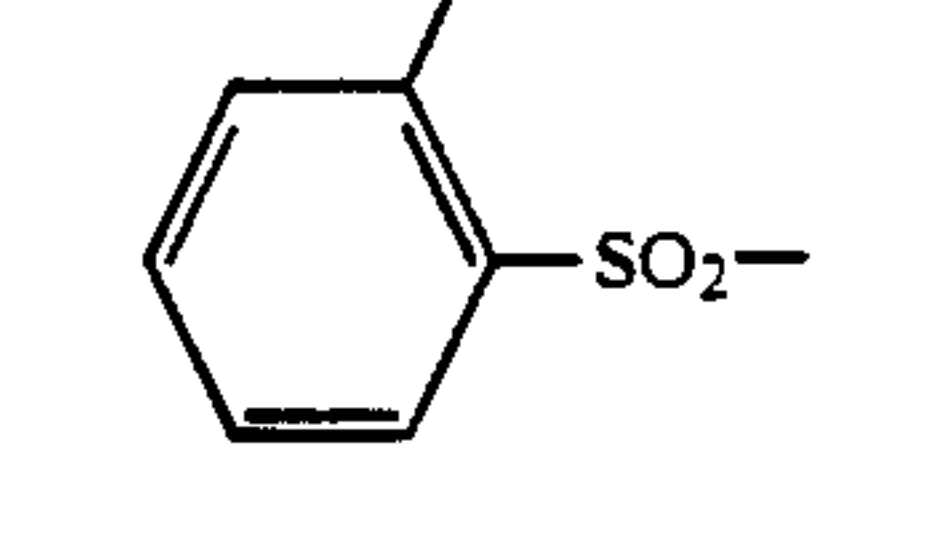
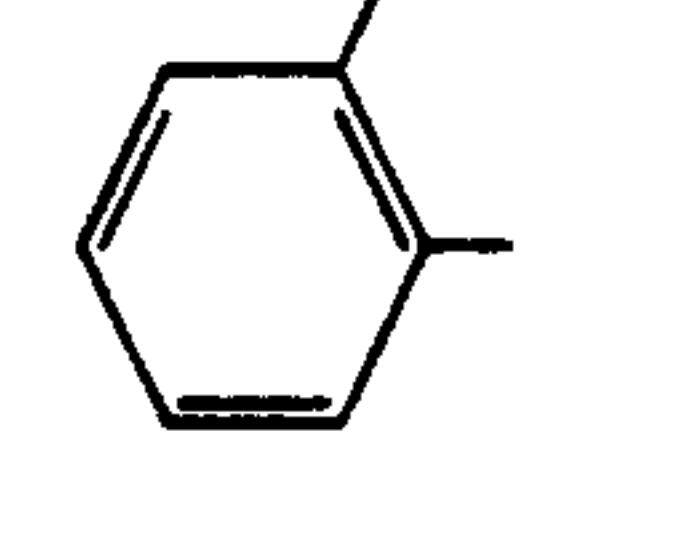
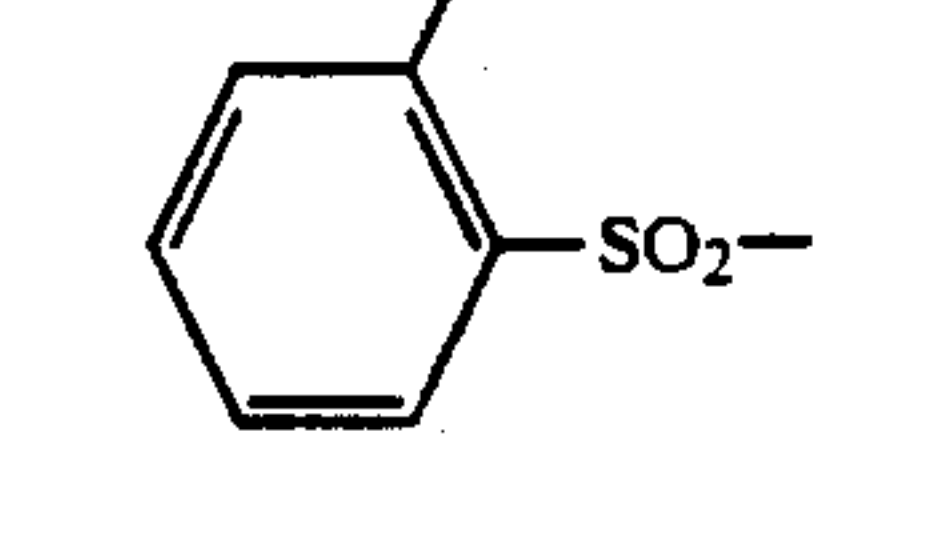
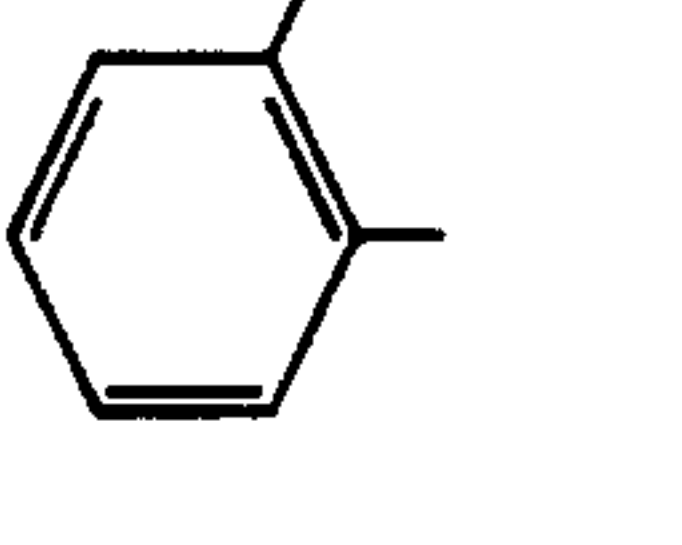
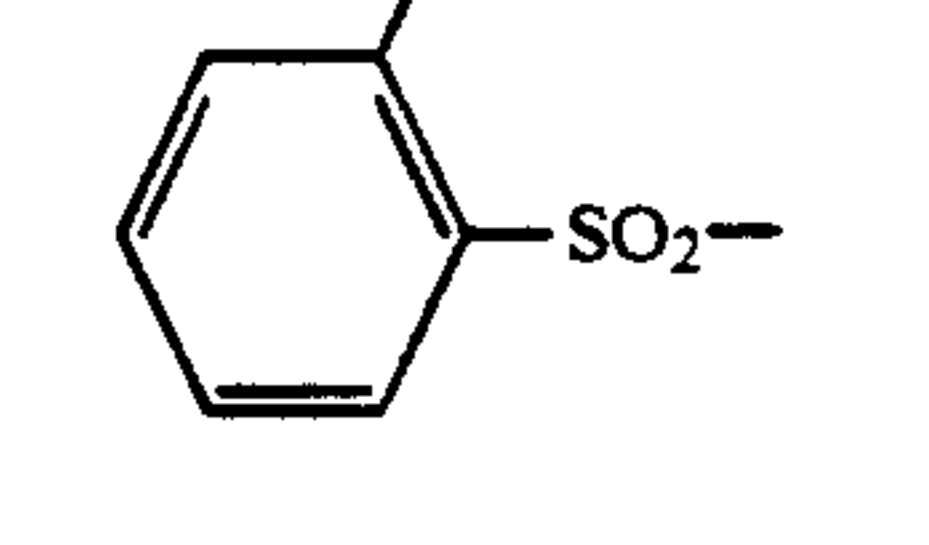
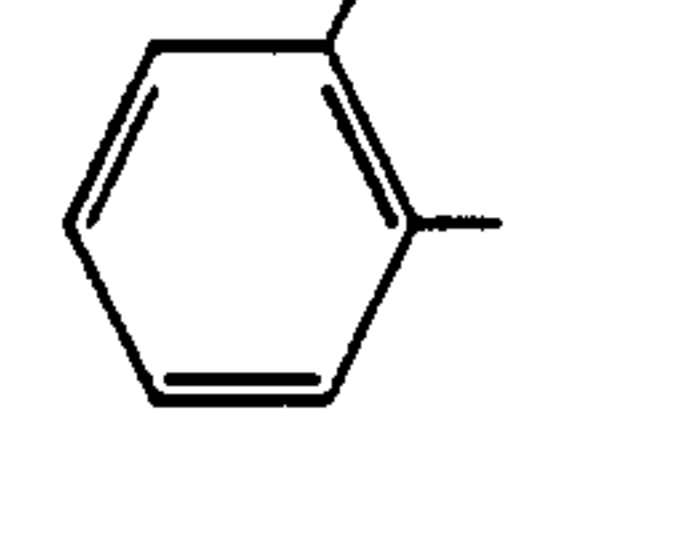
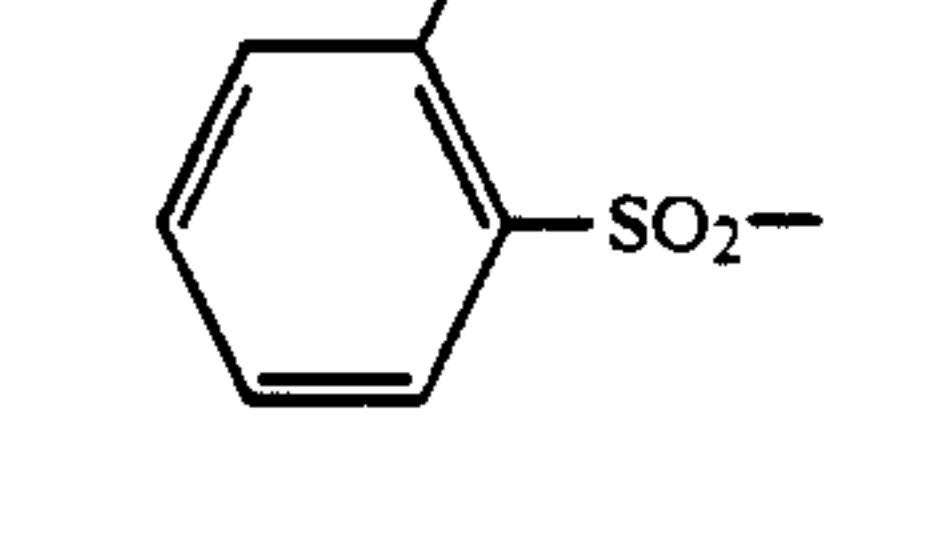
Ex- am- ple No.	Examples of the compounds of the formula (I)							Melting point (°C.)
	R ¹	R ²	R ³	R ⁴	X	Y	Z	
141			H	OCHF ₂	N	CH	C-OCHF ₂	
142			H	OCH ₃	N	CH	C-OCH ₃	
143			H	OCH ₃	N	CH	C-OCH ₃	
144			H	CH ₃	N	N	C-CH ₃	
145		H	H	CH ₃	N	N	C-CH ₃	
146			H	CH ₃	N	N	C-CH ₃	
147			H	OCH ₃	N	N	C-OCH ₃	
148			H	OCH ₃	N	N	C-OCH ₃	
149			H	OCH ₃	N	N	C-OCH ₃	
150			H	OCH ₃	N	N	C-Cl	

TABLE 2-continued

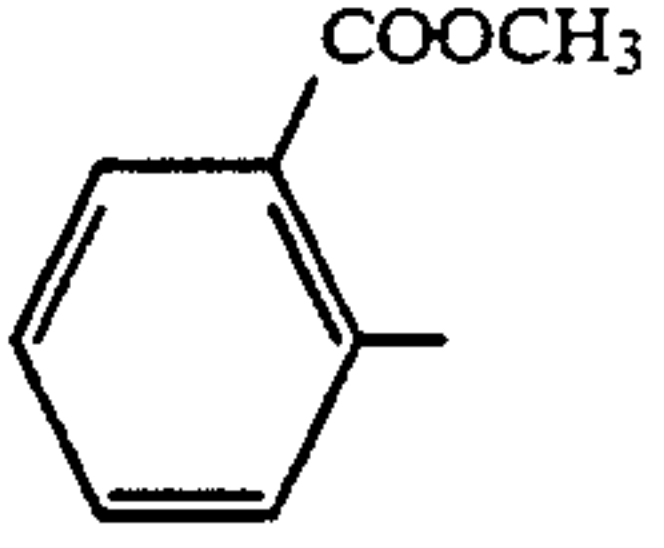
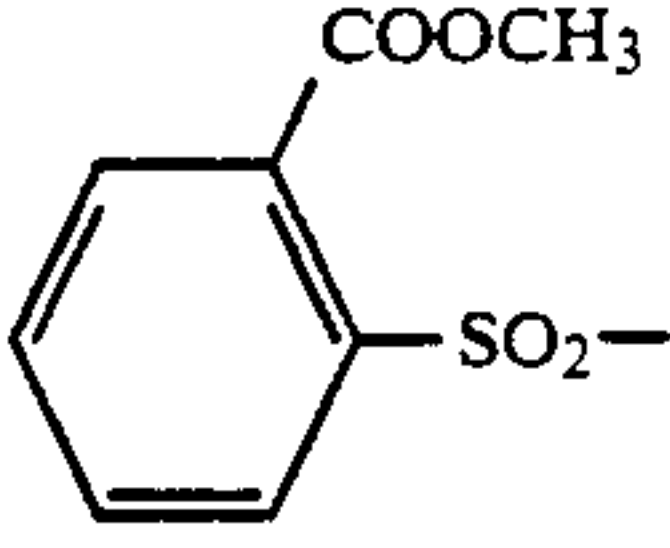
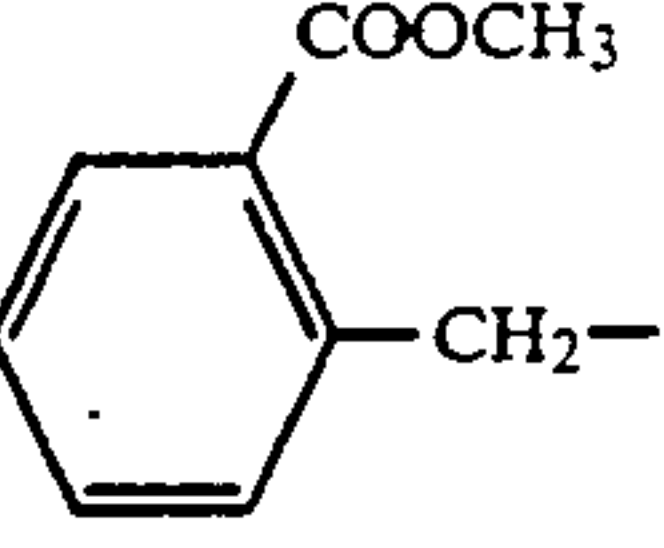
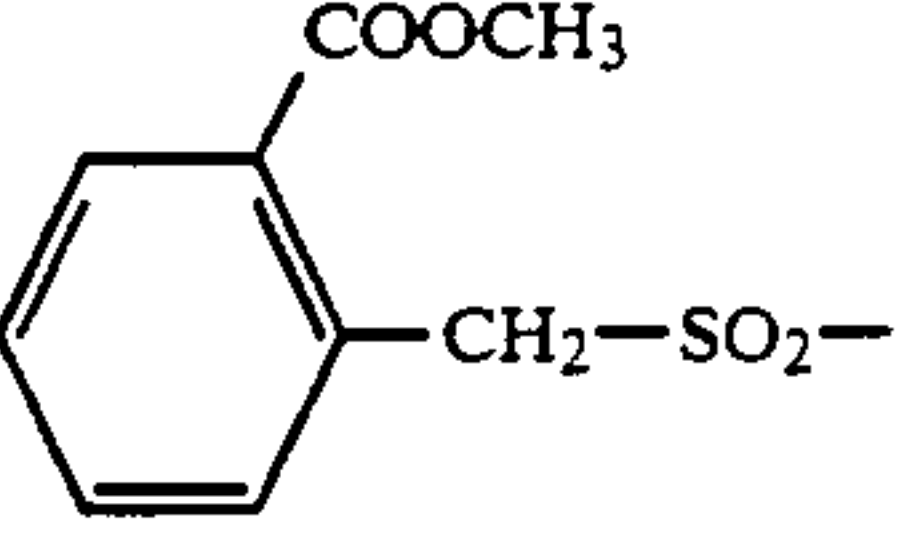
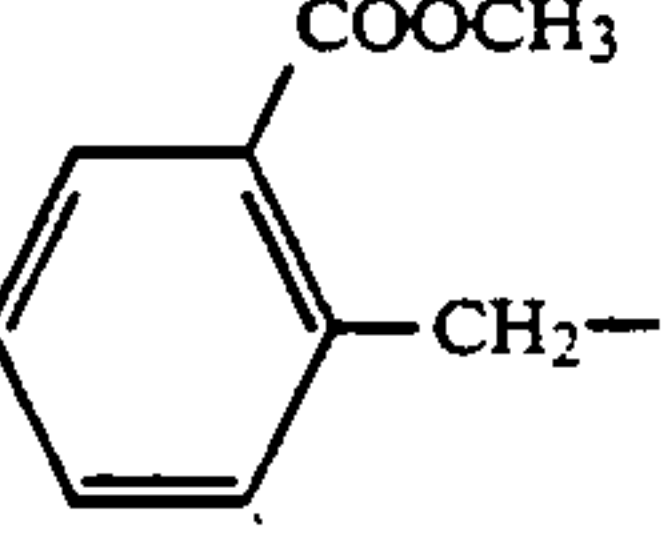
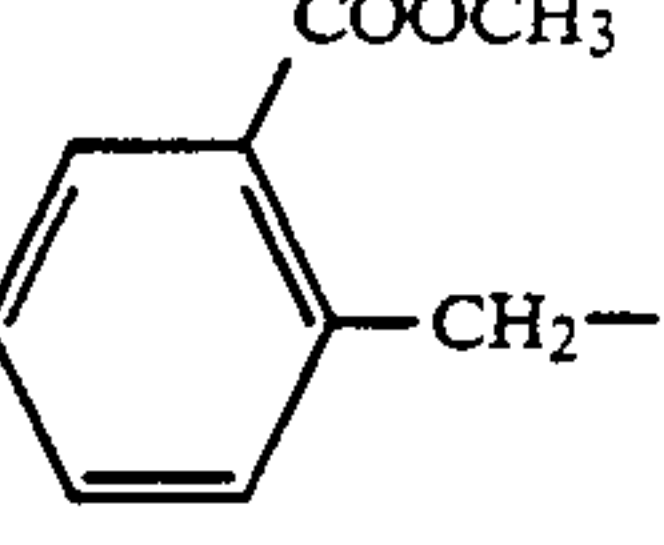
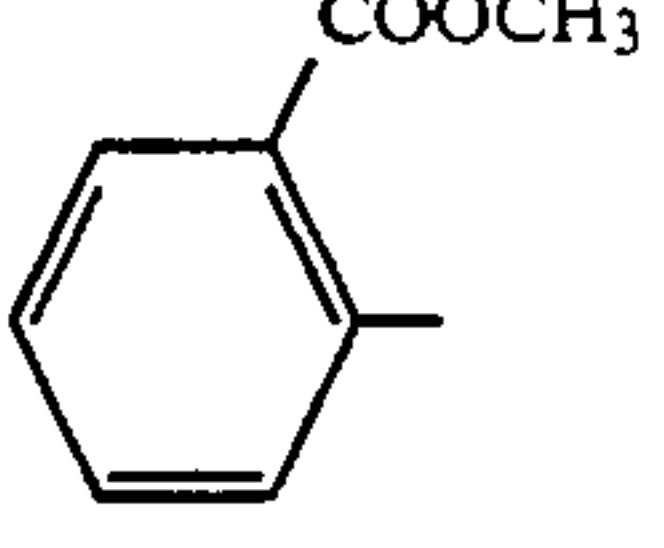
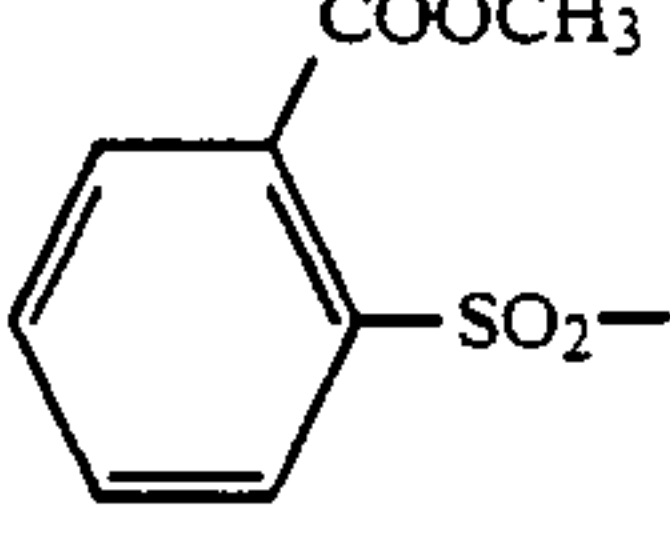
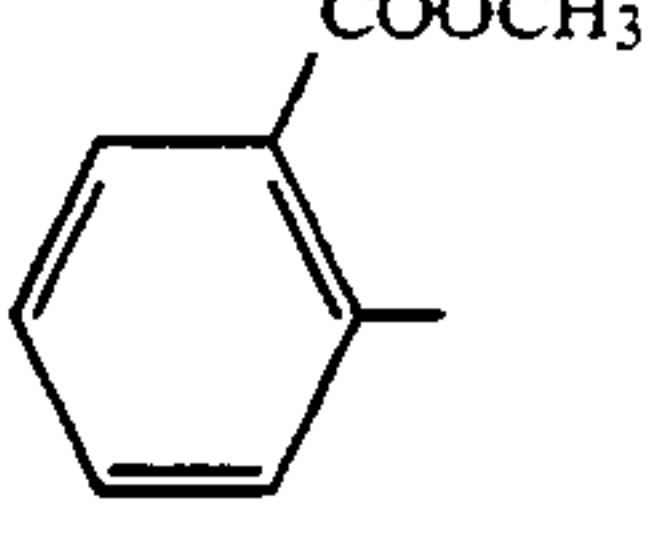
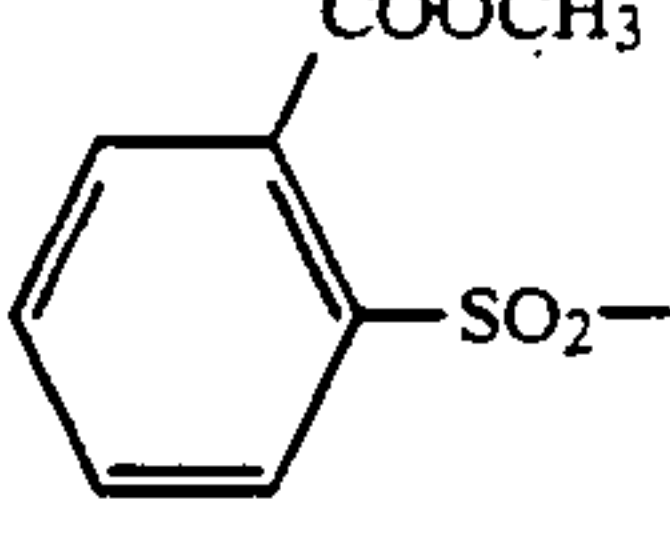
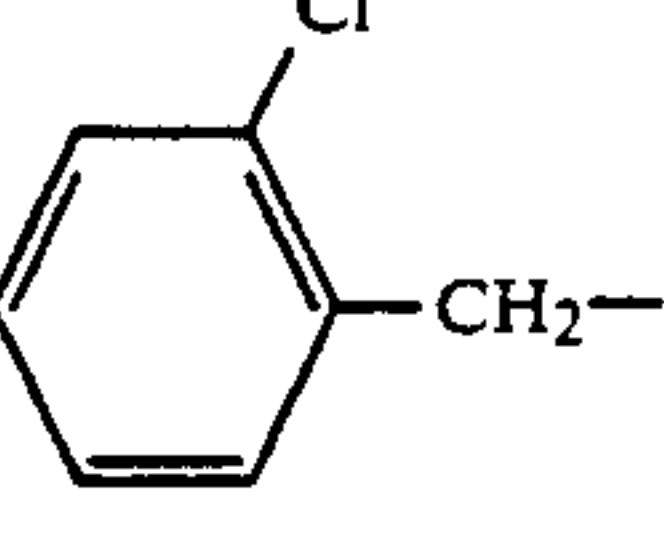
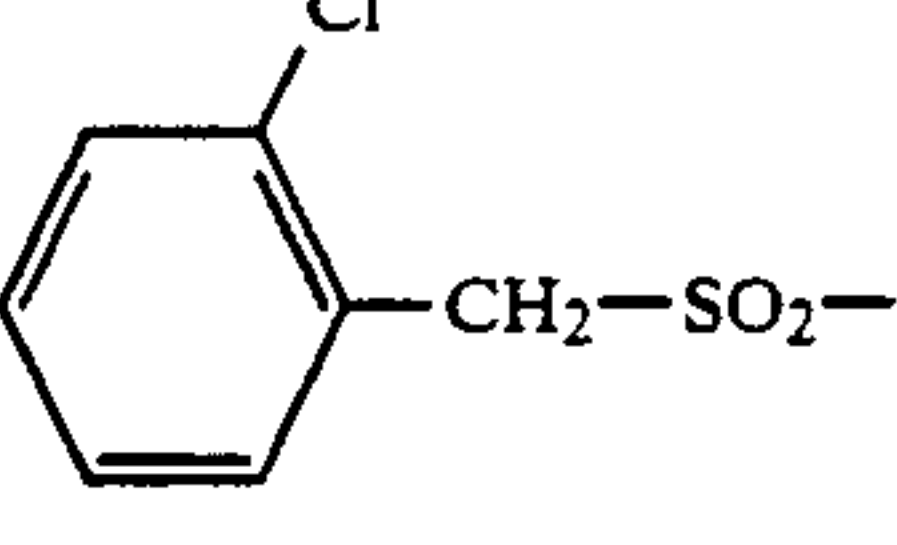
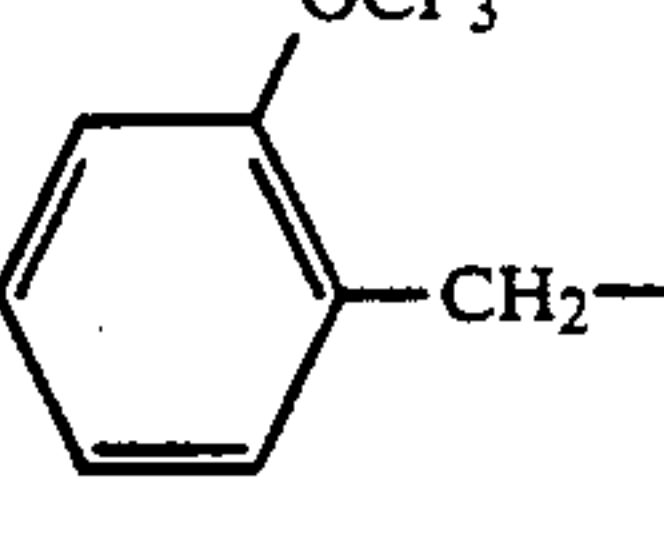
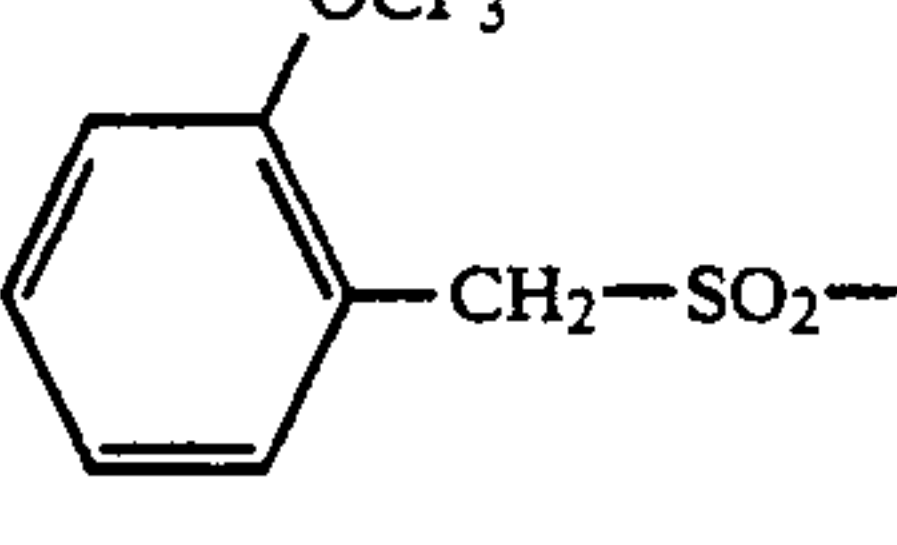
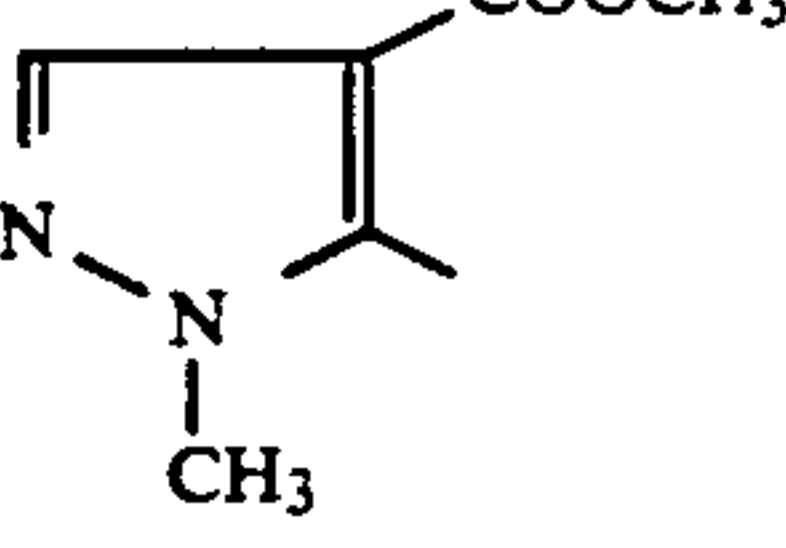
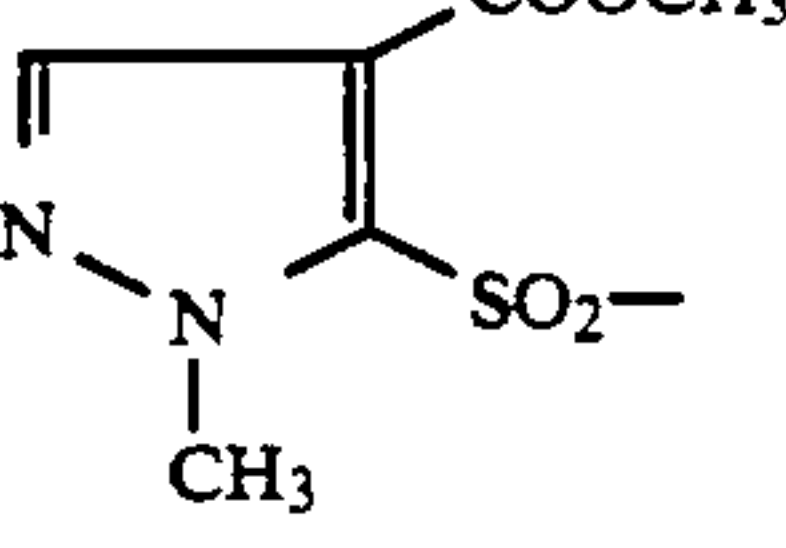
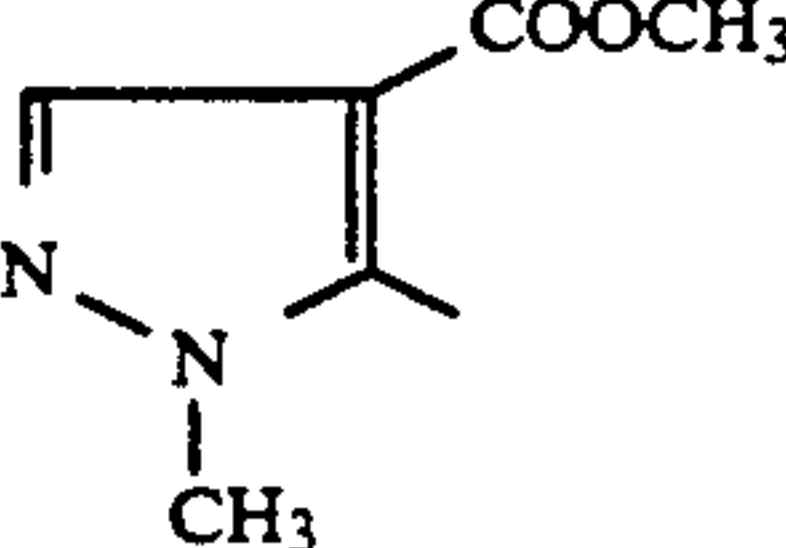
Ex- am- ple No.	Examples of the compounds of the formula (I)							Melting point (°C.)
	R ¹	R ²	R ³	R ⁴	X	Y	Z	
151			H	C ₂ H ₅	N	N	C-OCH ₃	
152			H	OCH ₃	N	N	C-OCH ₃	
153		H	H	OCH ₃	N	N	C-OCH ₃	
154		H	H	OCH ₃	N	N	C-CH ₃	
155			H	OCH ₃	N	N	C-NHC ₂ H ₅	
156			H	OC ₂ H ₅	N	N	C-NHCH ₃	
157			H	OCH ₃	N	N	C-OCH ₃	
158			H	OCH ₃	N	N	C-OCH ₃	
159			H	OCH ₃	N	CH	C-OCH ₃	
160		H	H	OCH ₃	N	CH	C-OCH ₃	

TABLE 2-continued

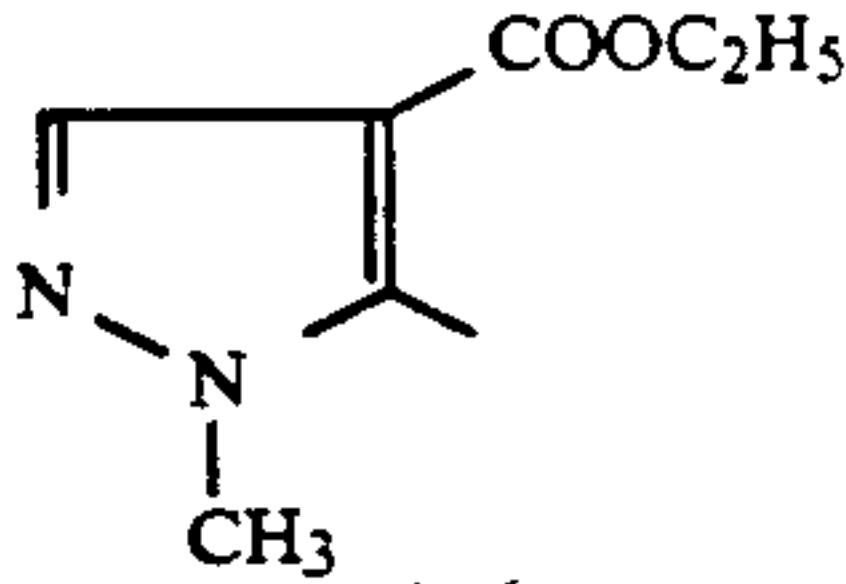
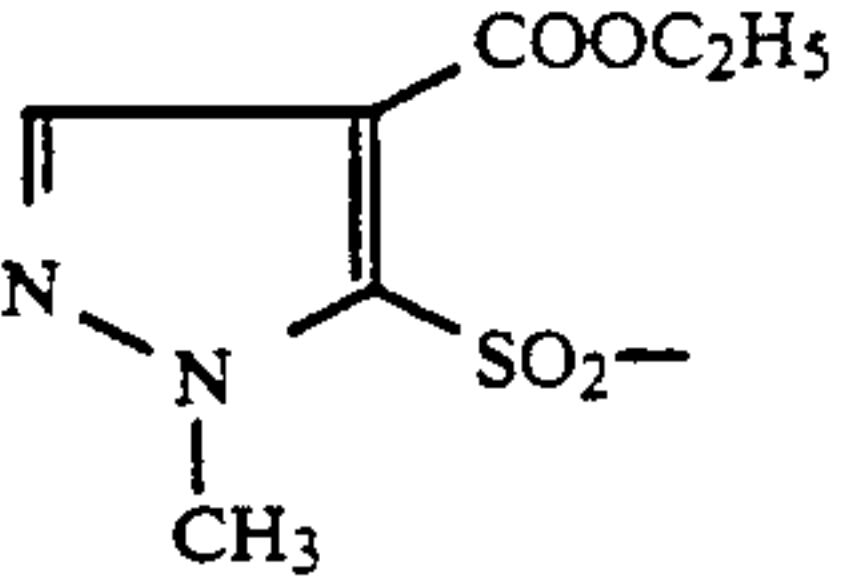
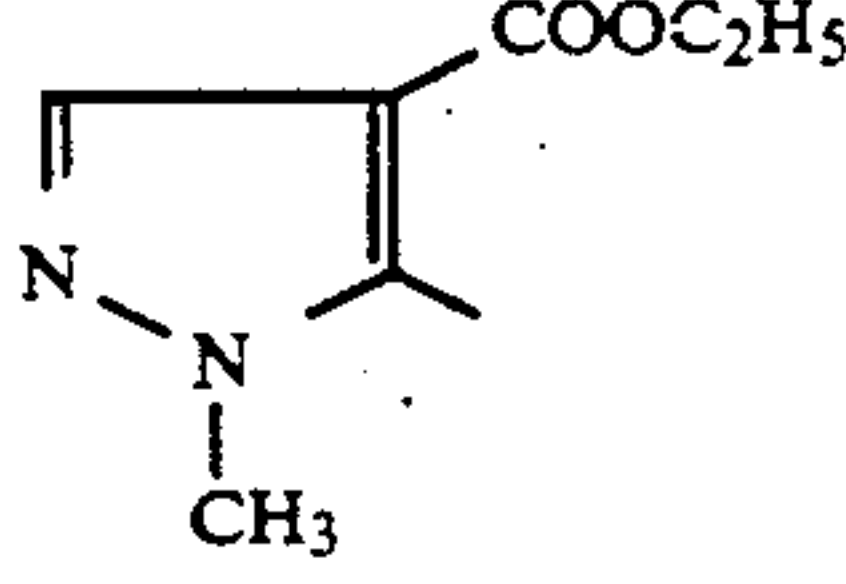
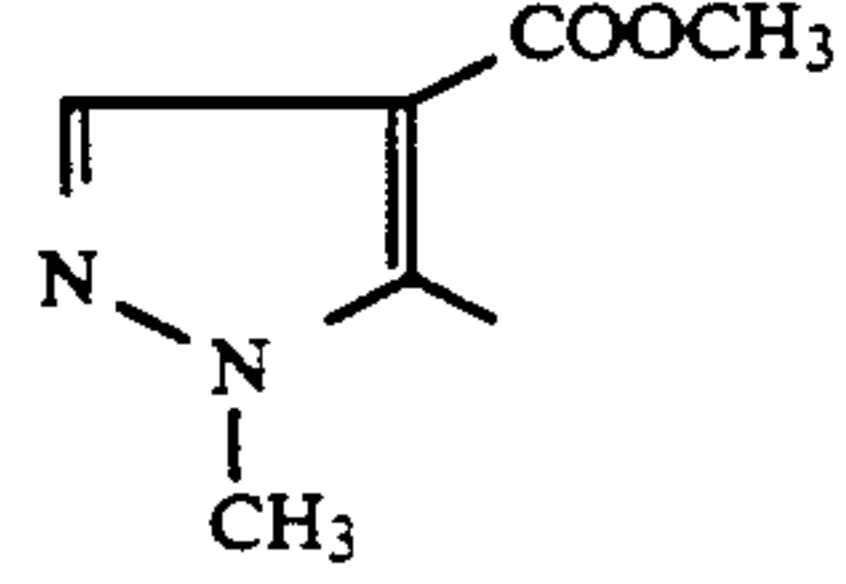
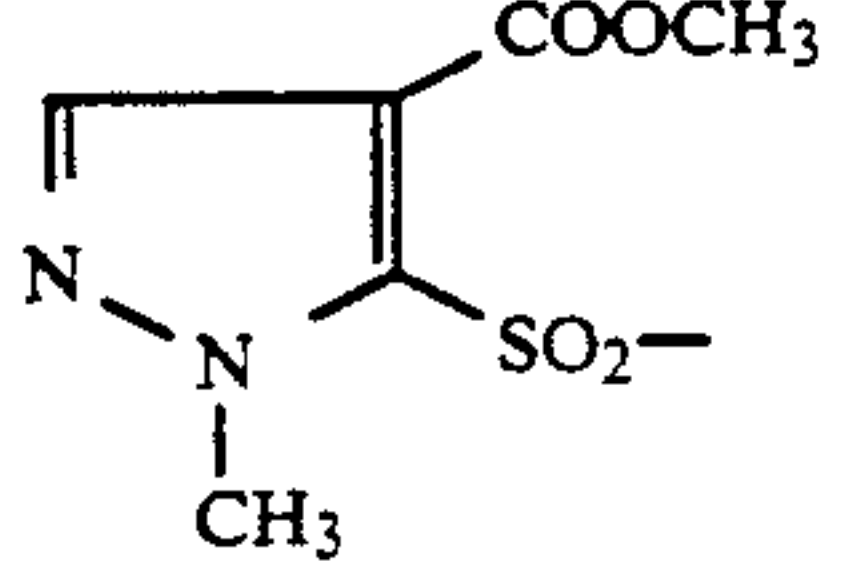
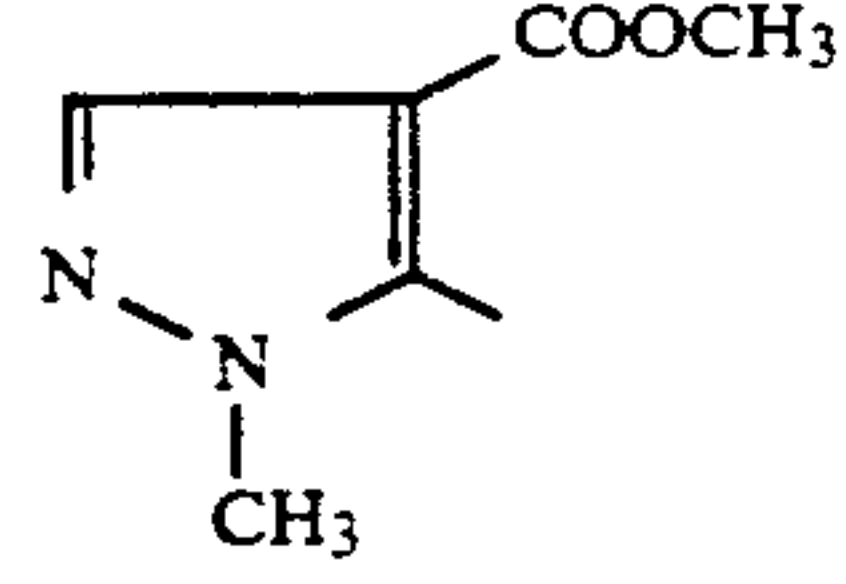
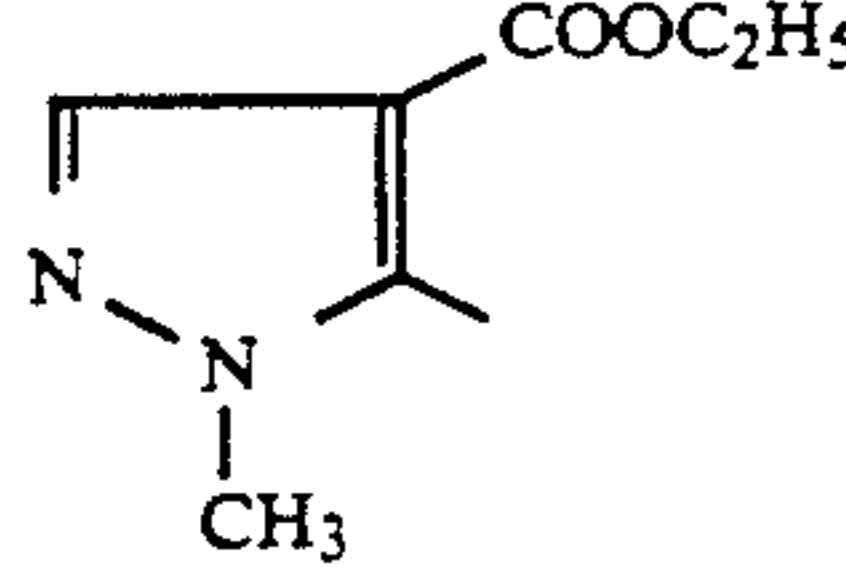
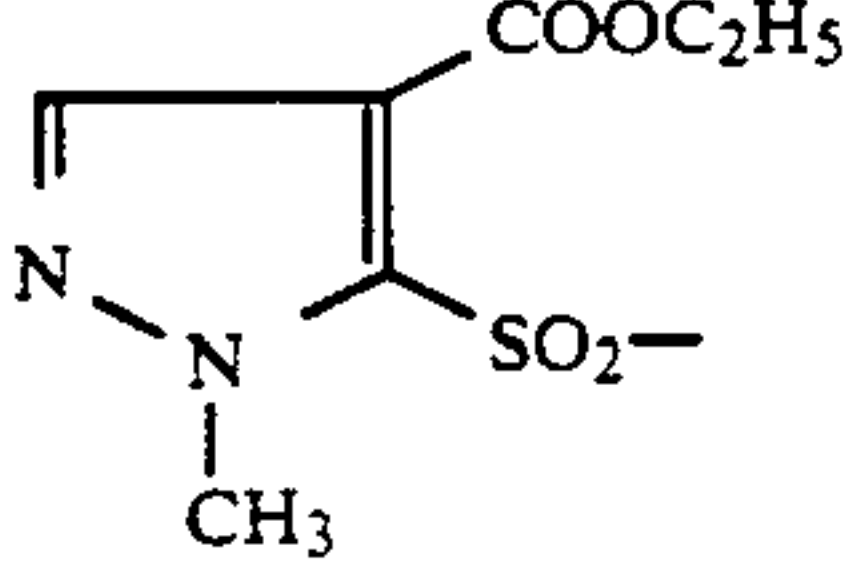
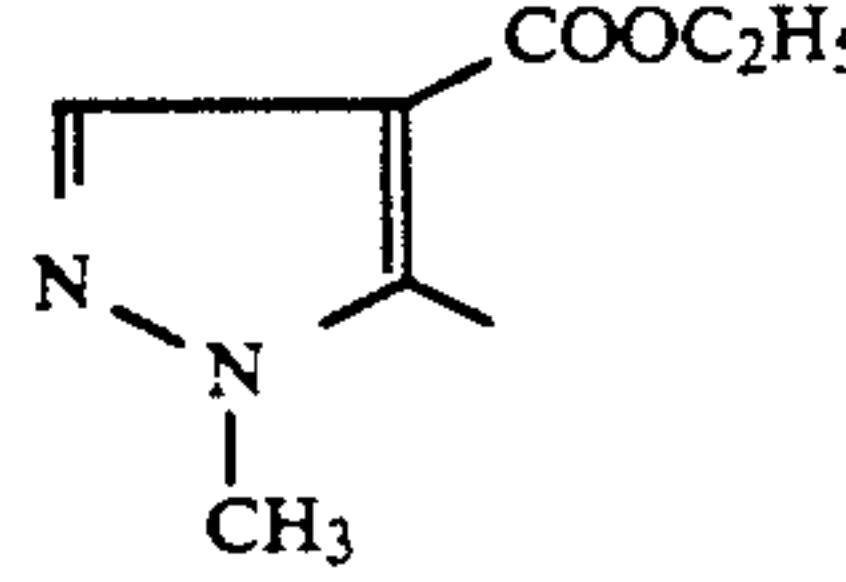
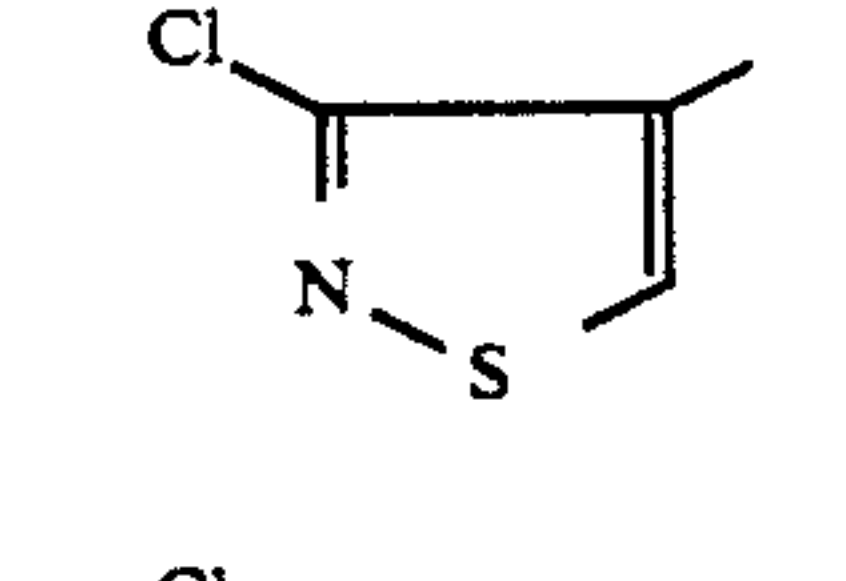
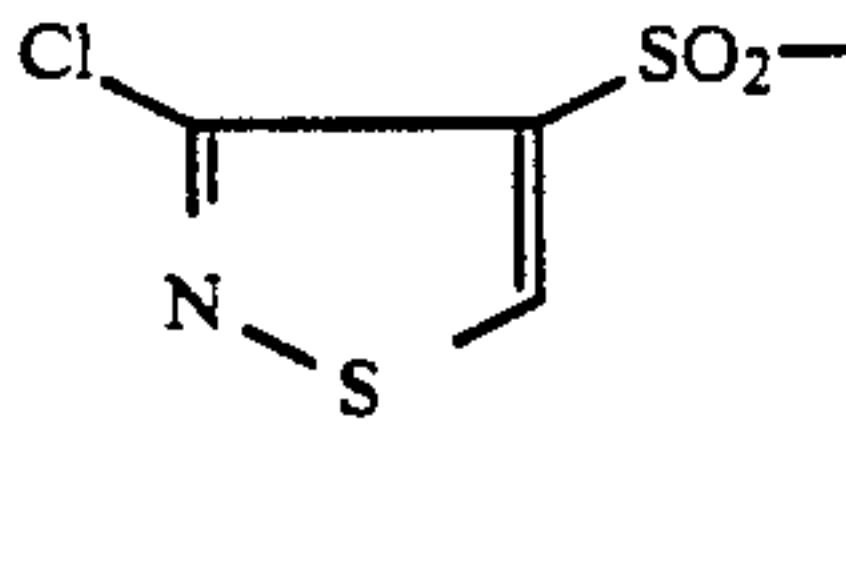
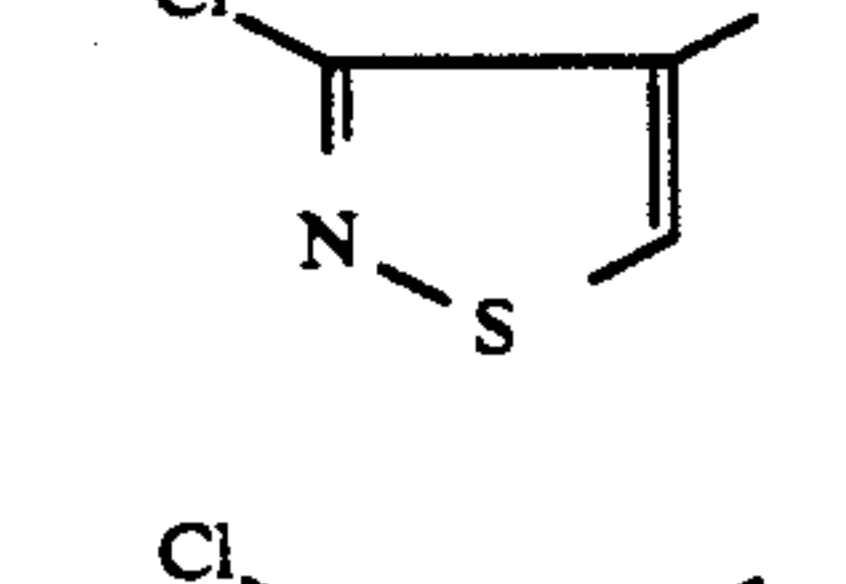
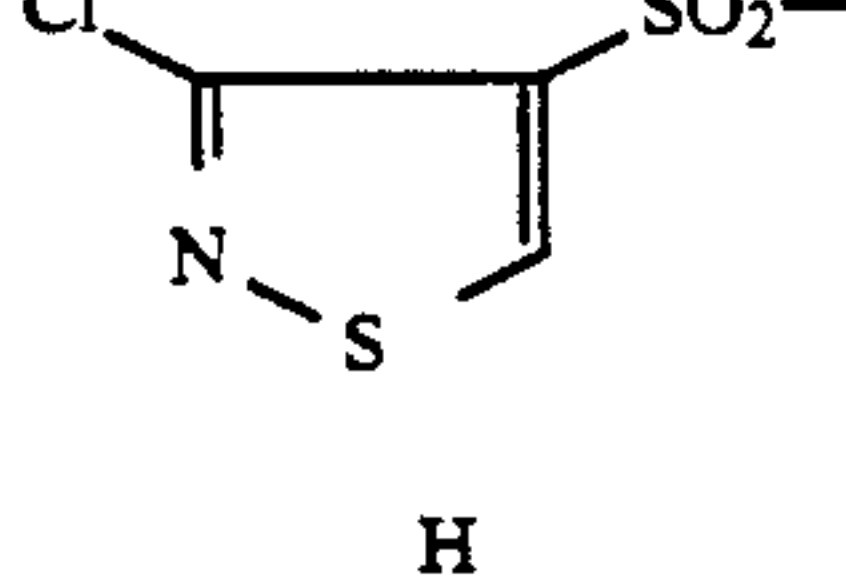
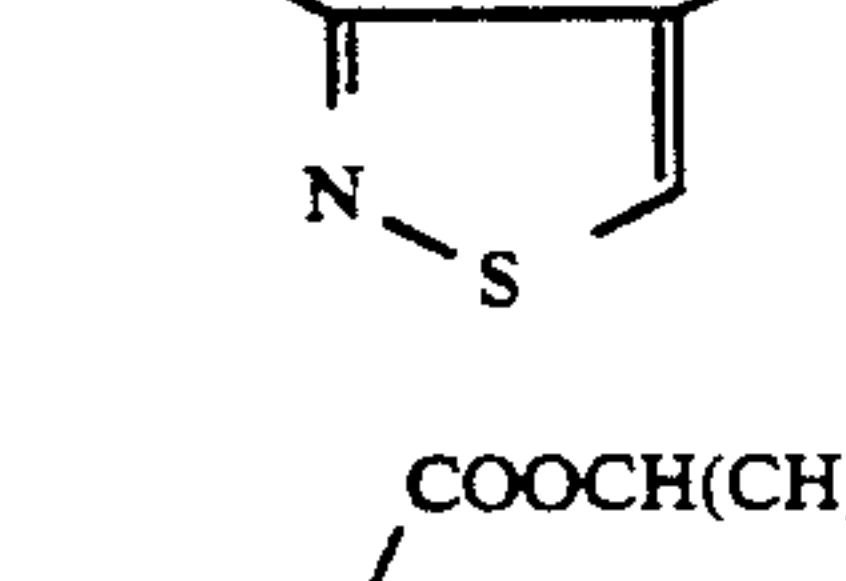
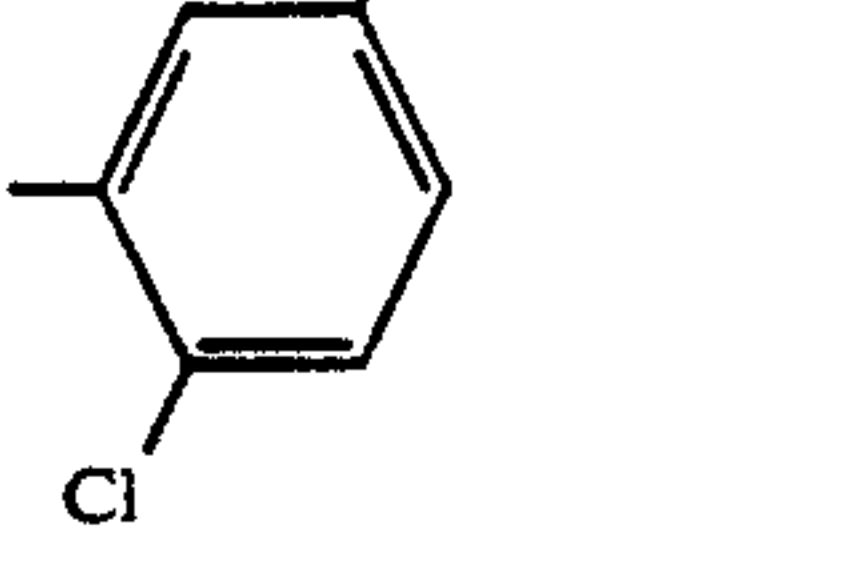
Ex- am- ple No.	Examples of the compounds of the formula (I)							Melting point (°C.)
	R ¹	R ²	R ³	R ⁴	X	Y	Z	
161			H	OCH ₃	N	CH	C—OCH ₃	
162		H	H	OCH ₃	N	CH	C—OCH ₃	
163			H	OCH ₃	N	N	C—OCH ₃	
164		H	H	OCH ₃	N	N	C—OCH ₃	
165			H	OCH ₃	N	N	C—OCH ₃	
166		H	H	OCH ₃	N	N	C—OCH ₃	
167			H	OCH ₃	N	N	C—OCH ₃	
168			H	CH ₃	N	CH	C—CH ₃	
169		H	H	CH ₃	N	CH	C—CH ₃	
170		H	H	OCH ₃	N	N	C—OCH ₃	

TABLE 2-continued

Examples of the compounds of the formula (I)

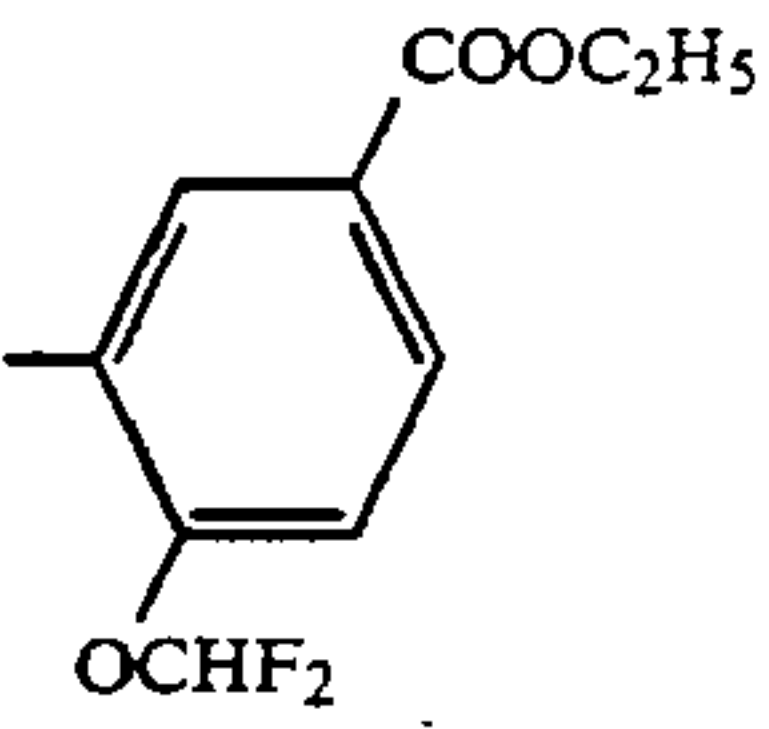
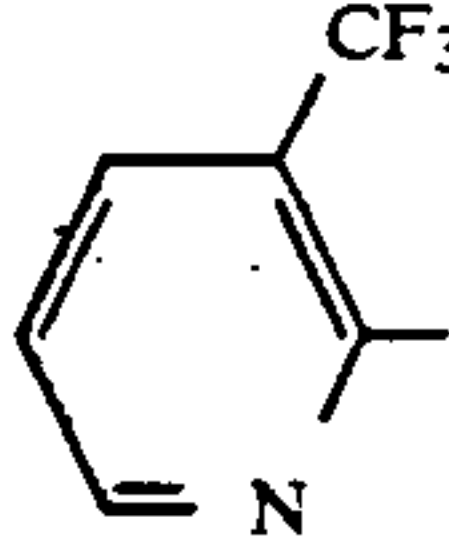
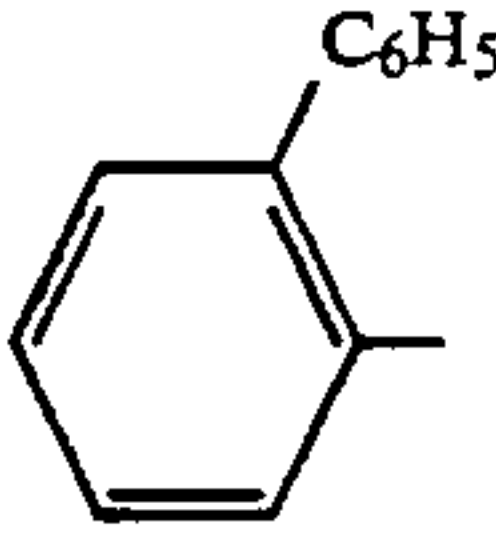
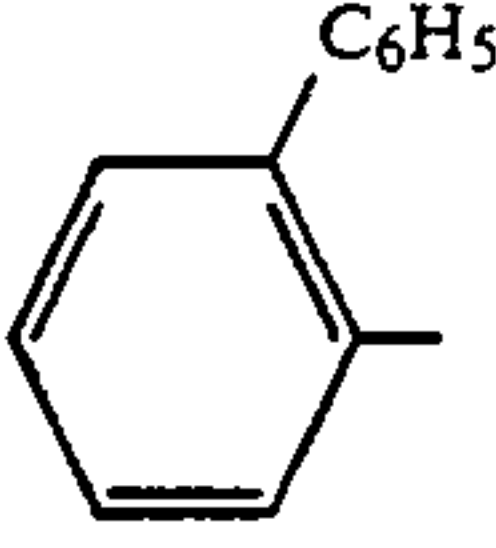
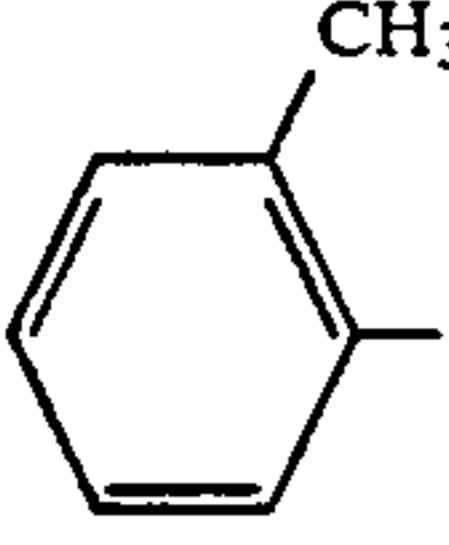
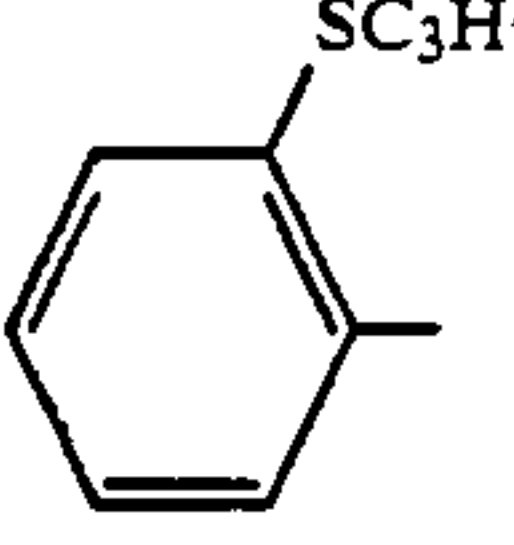
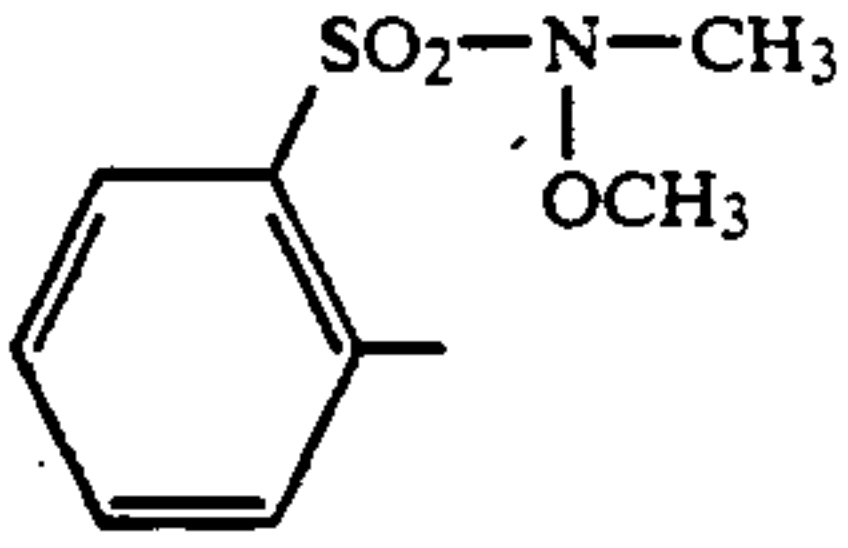
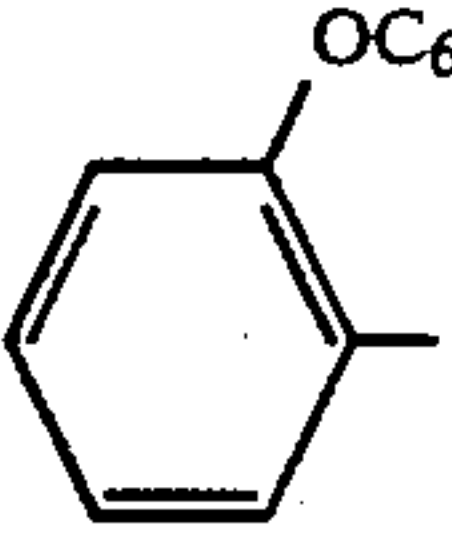
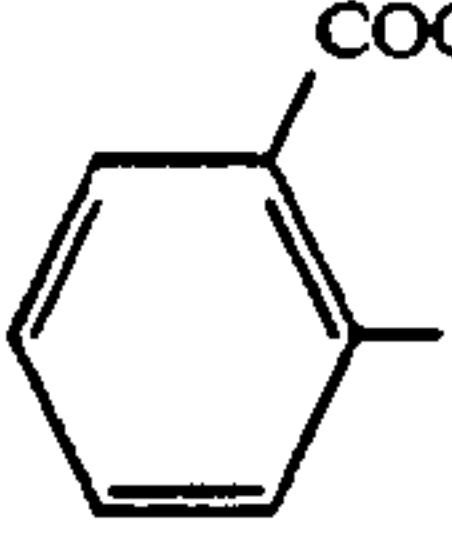
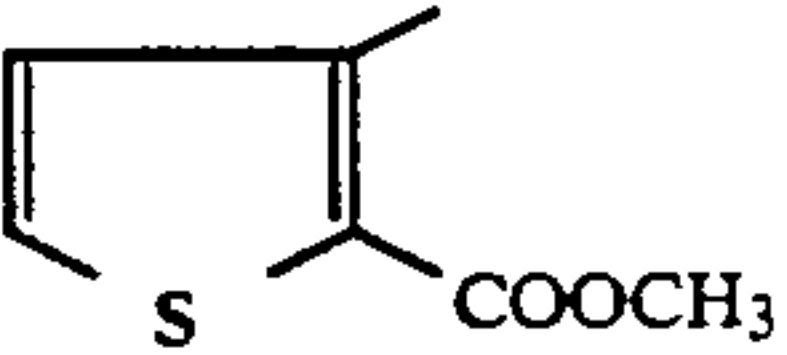
Ex- am- ple No.	R ¹	R ²	R ³	R ⁴	X	Y	Z	Melting point (°C.)
171		H	H	OCH ₃	N	CH	C—OCH ₃	
172		H	H	OCH ₃	N	CH	C—OCH ₃	
173		H	H	OCH ₃	N	CH	C—OCH ₃	
174		H	H	OCH ₃	N	N	C—OCH ₃	
175		H	H	OCH ₃	N	N	C—OCH ₃	
176		H	H	OCH ₃	N	N	C—OCH ₃	
177		H	H	OCH ₃	N	N	C—OCH ₃	
178		H	H	OCH ₃	N	N	C—OCH ₃	
179		H	CH ₃	CH ₃	N	CH	C—CH ₃	233
180		H	CH ₃	CH ₃	N	CH	C—CH ₃	

TABLE 2-continued

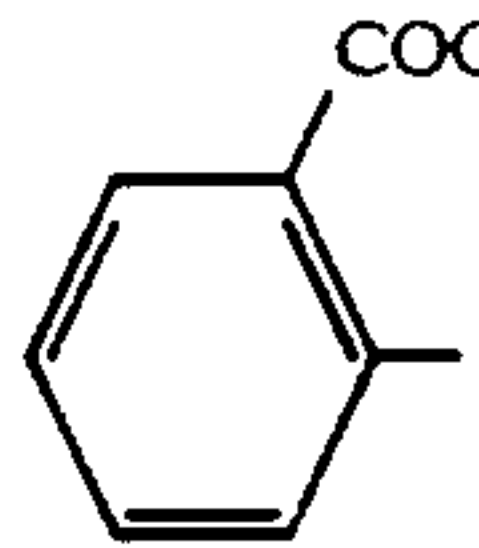
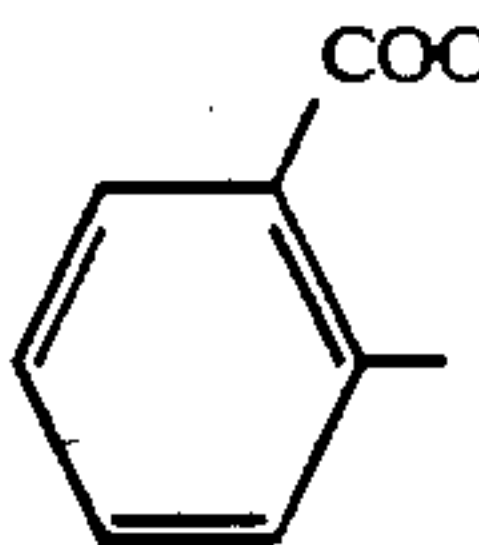
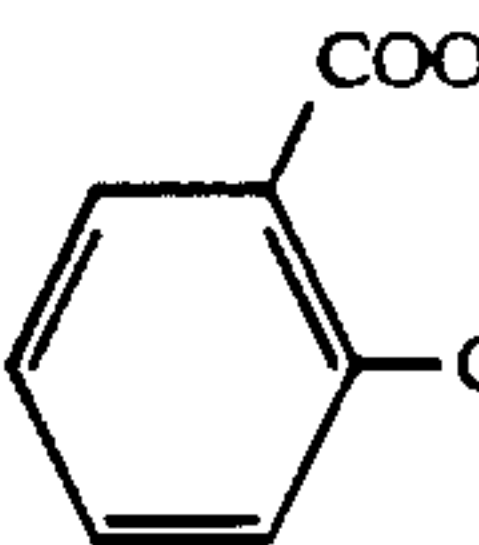
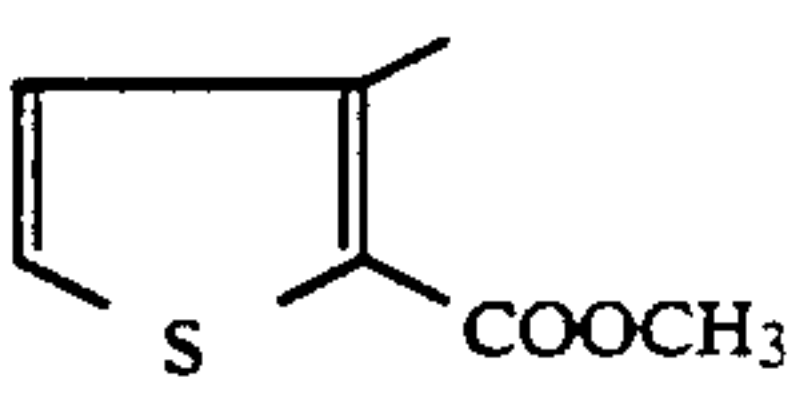
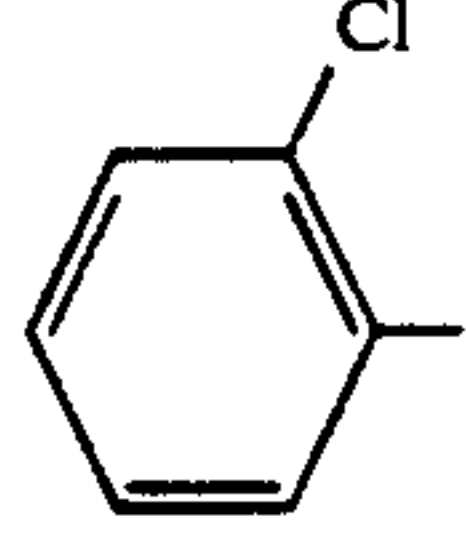
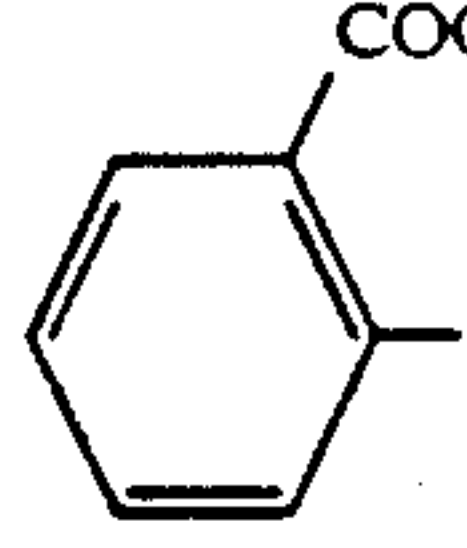
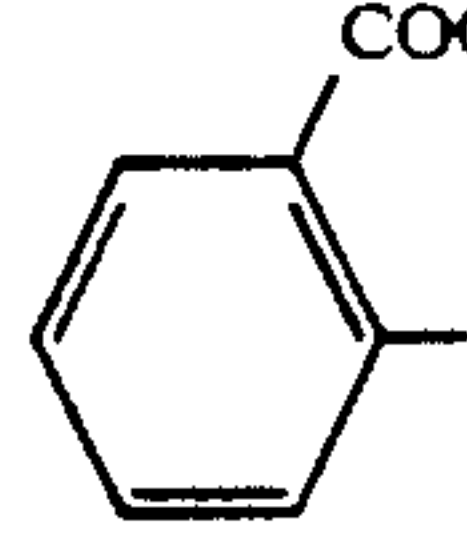
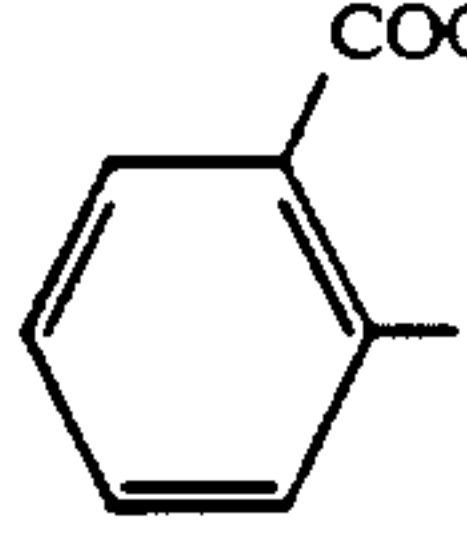
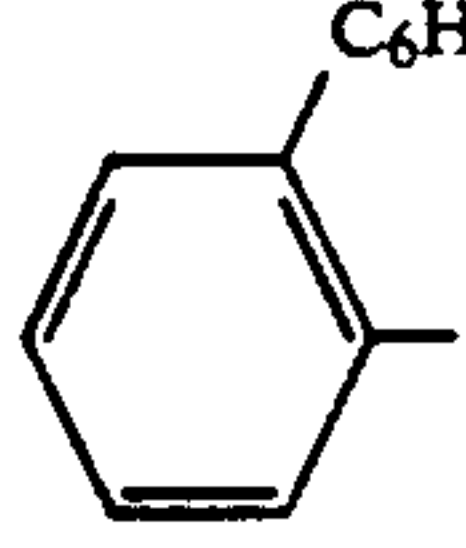
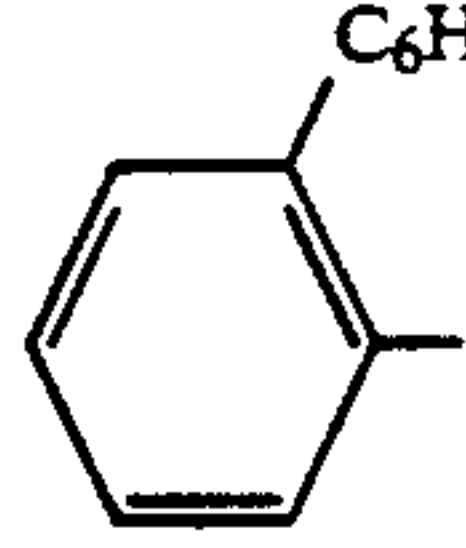
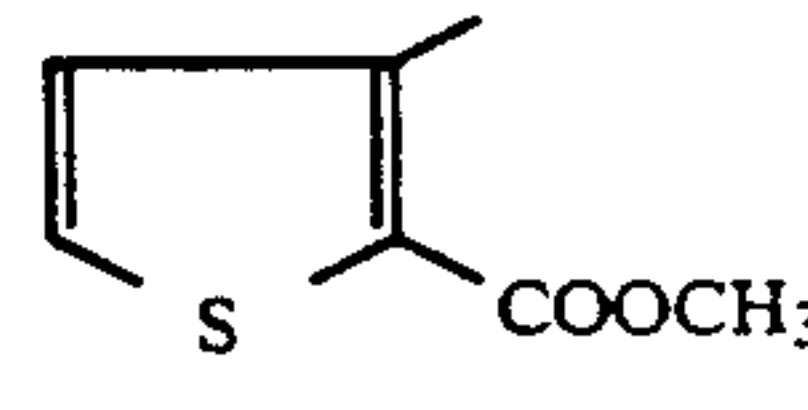
Examples of the compounds of the formula (I)								Melting point (°C.)
Ex-ample No.	R ¹	R ²	R ³	R ⁴	X	Y	Z	
181		H	SCH ₃	CH ₃	N	CH	C—CH ₃	
182		H	CH ₃	CH ₃	N	CH	C—CH ₃	
183		H	SCH ₃	OCH ₃	N	CH	C—OCH ₃	
184		H	CH ₃	OCH ₃	N	N	C—OCH ₃	
185		H	CH ₃	CH ₃	N	N	C—OCH ₃	
186		H	CH ₃	CH ₃	N	N	C—OCH ₃	
187		H	CH ₃	OCH ₃	N	N	C—OCH ₃	202
188		H	CH ₃	OCH ₃	N	CH	C—Cl	
189		H	CH ₃	OCH ₃	N	CH	C—OCH ₃	
190		H	CH ₃	OCH ₃	N	N	C—OCH ₃	
191		H	CH ₃	CH ₃	N	N	C—OCH ₃	

TABLE 2-continued

Ex- am- ple No.	Examples of the compounds of the formula (I)							Melting point (°C.)
	R ¹	R ²	R ³	R ⁴	X	Y	Z	
192			H	CH ₃	N	CH	C-CH ₃	
193		H	H	CH ₃	N	CH	C-CH ₃	
194		H	SCH ₃	CH ₃	N	N	C-OCH ₃	
195		H	SCH ₃	CH ₃	N	N	C-OCH ₃	
196		H	SCH ₃	OCH ₃	N	N	C-OCH ₃	
197		H	H	SO ₂ CH ₃	CH	N	C-OCH ₃	
198		H	CH ₃	SO ₂ CH ₃	CH	N	C-OCH ₃	
199			CH ₃	OCHF ₂	CH	N	C-OCHF ₂	200
200			C ₂ H ₅	CH ₃	N	N	C-OCH ₃	
201		H	CH(CH ₃) ₂	OCH ₃	N	CH	C-OCH ₃	
202		H	CH ₂ CH(CH ₃) ₂	OCH ₃	N	N	C-OCH ₃	

TABLE 2-continued

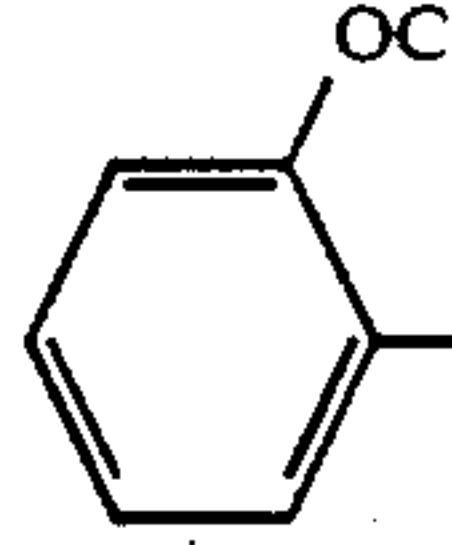
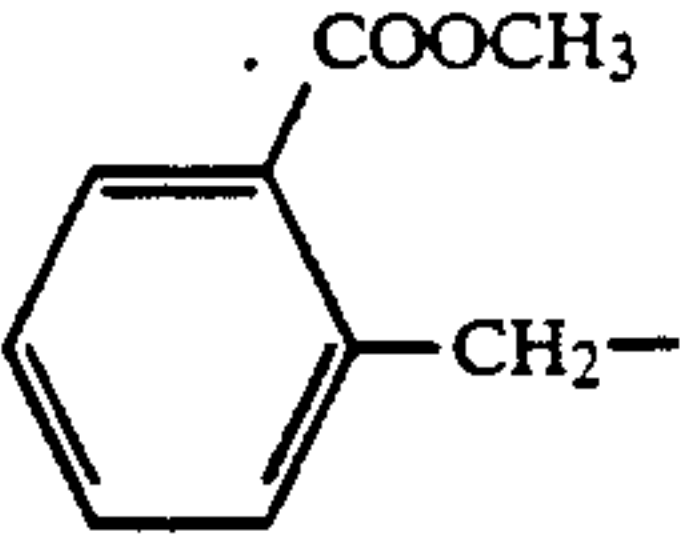
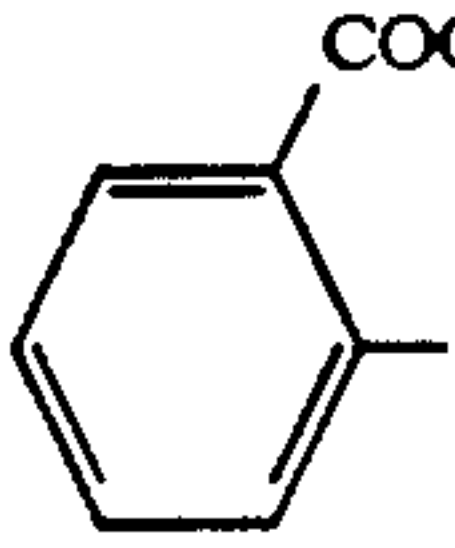
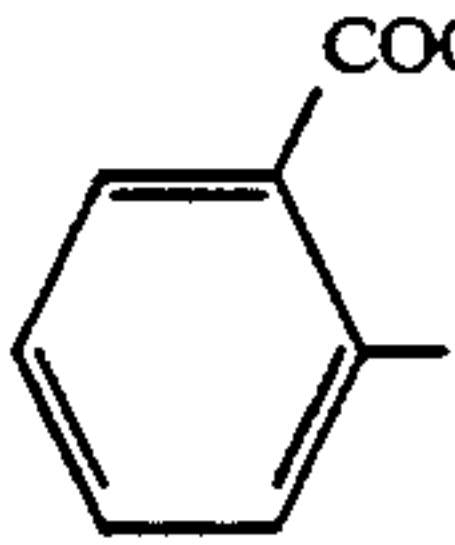
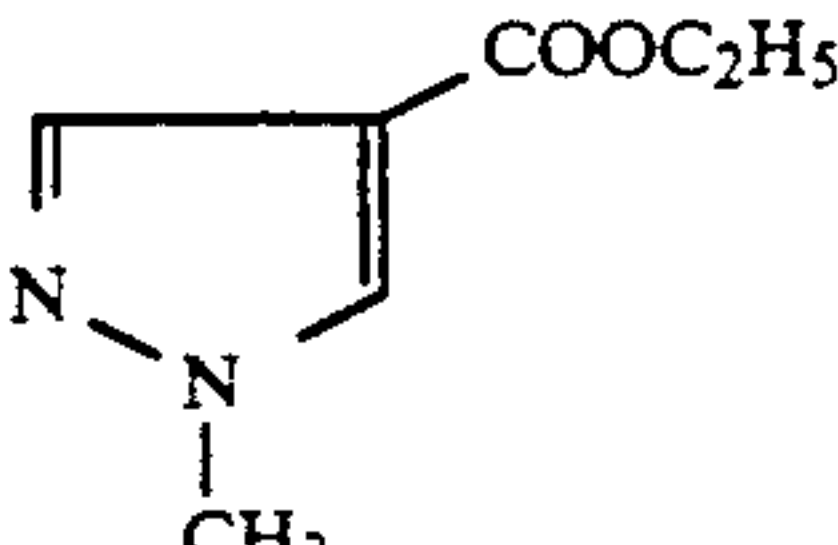
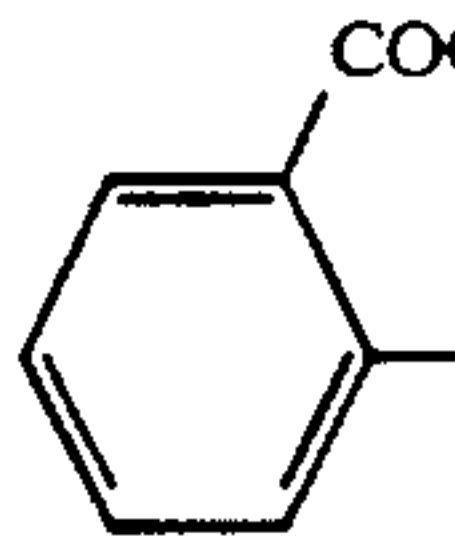
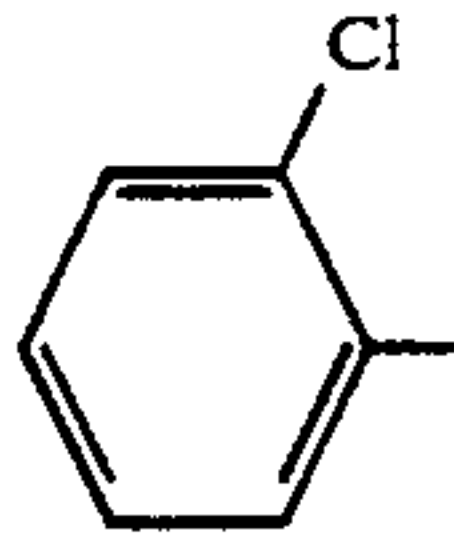
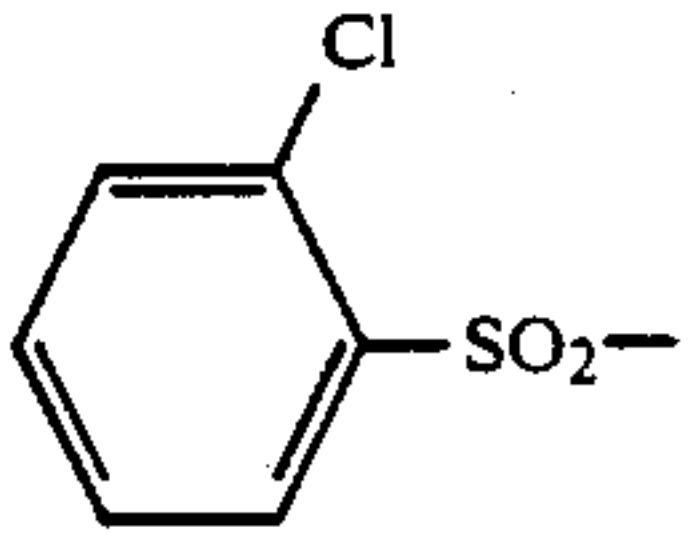
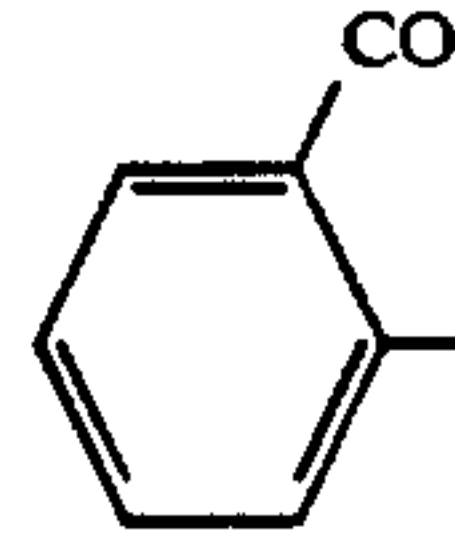
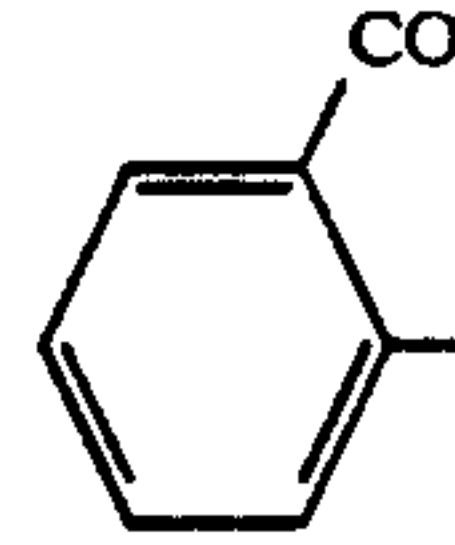
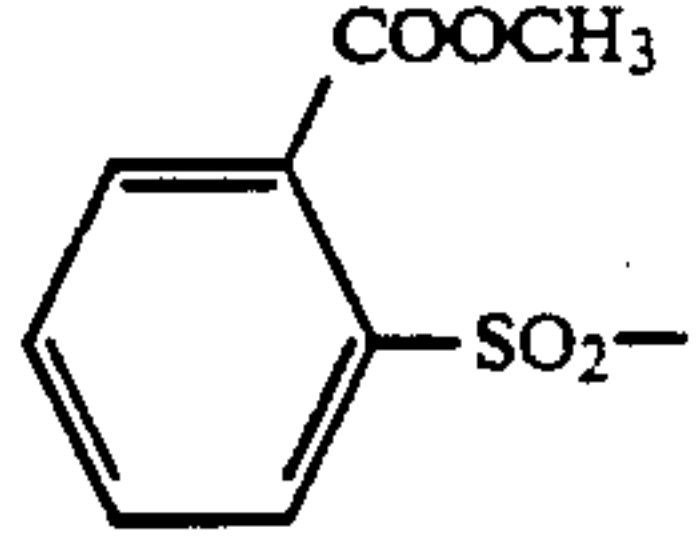
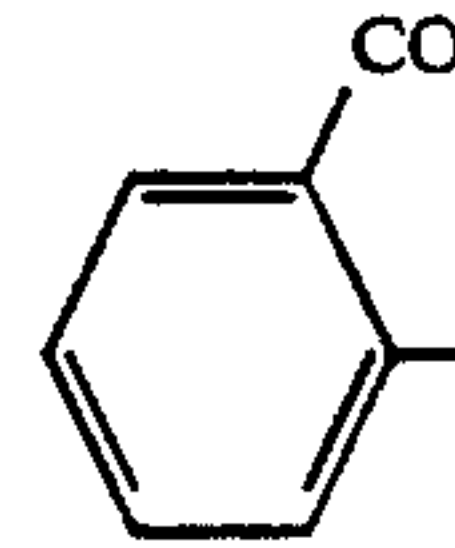
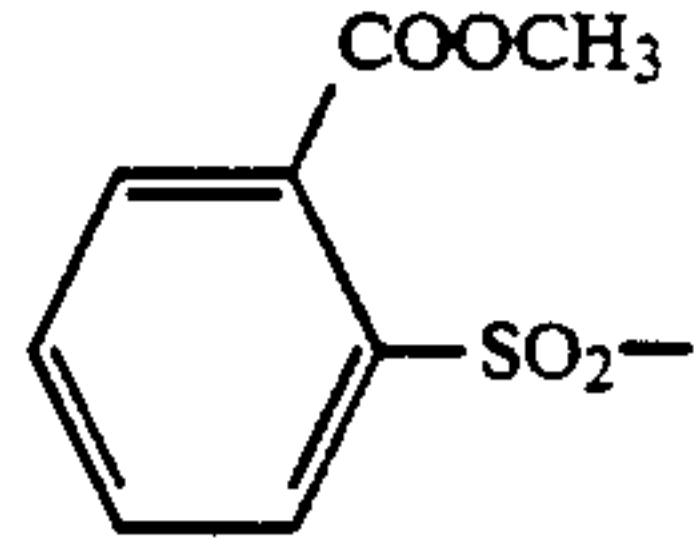
Examples of the compounds of the formula (I)								
Ex- am- ple No.	R ¹	R ²	R ³	R ⁴	X	Y	Z	Melting point (°C.)
203		H	C(CH ₃) ₃	CH ₃	N	CH	C—OCH ₃	
204		H	C(CH ₃) ₃	OCH ₃	N	CH	C—OCH ₃	
205		H	CF ₃	CH ₃	N	N	C—OCH ₃	
206		H	CH ₃	OCHF ₂	N	CH	C—OCHF ₂	118
207		H	CH ₃	OCH ₃	N	CH	C—OCH ₃	198
208		H	C ₂ H ₅	OCH ₃	N	CH	C—OCH ₃	174
209			CH ₃	OCH ₃	N	CH	C—OCH ₃	204
210		H	C ₃ H ₇	OCH ₃	N	CH	C—OCH ₃	179
211			CH(CH ₃) ₂	OCH ₃	N	CH	C—OCH ₃	184
212			CH ₃	CH ₃	N	CH	C—OCH ₃	

TABLE 2-continued

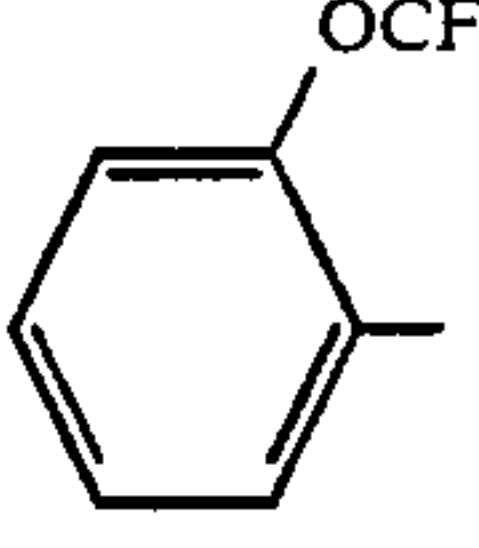
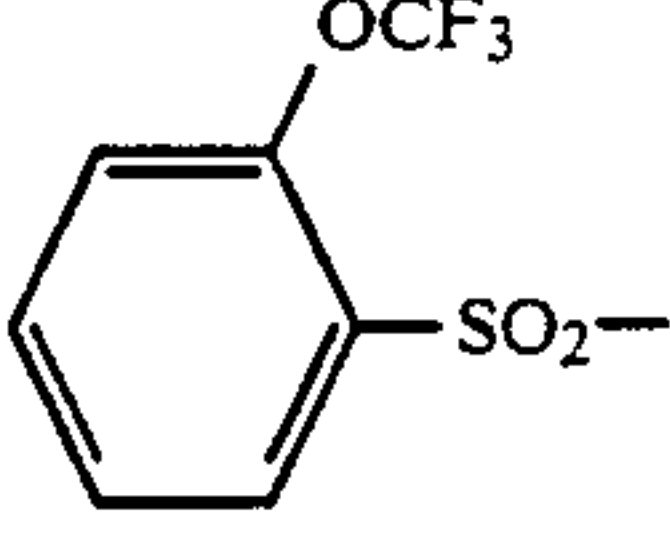
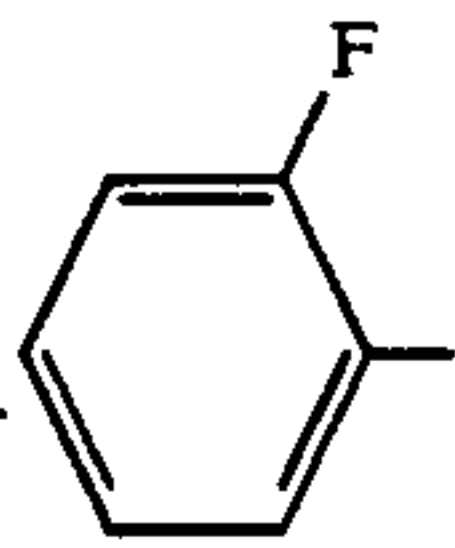
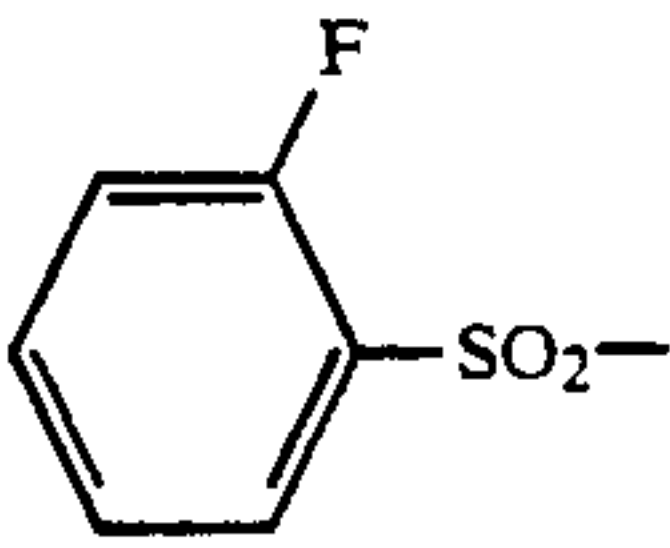
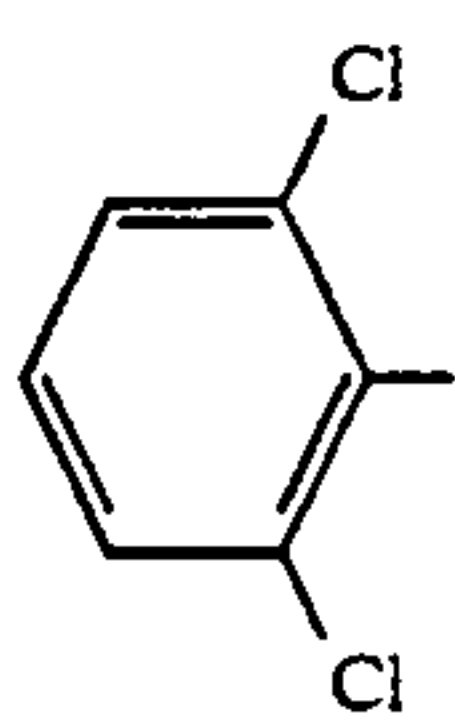
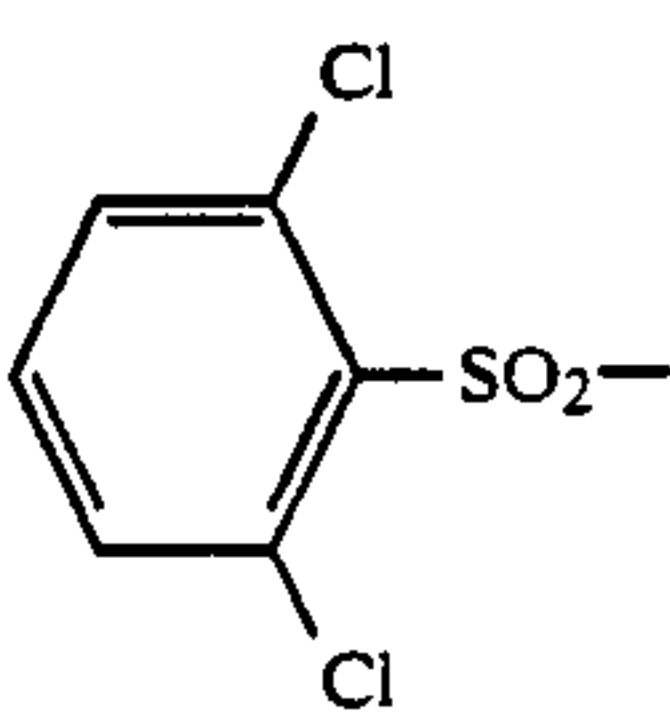
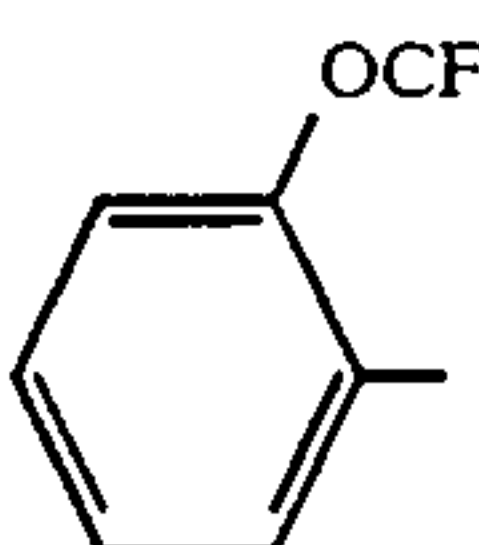
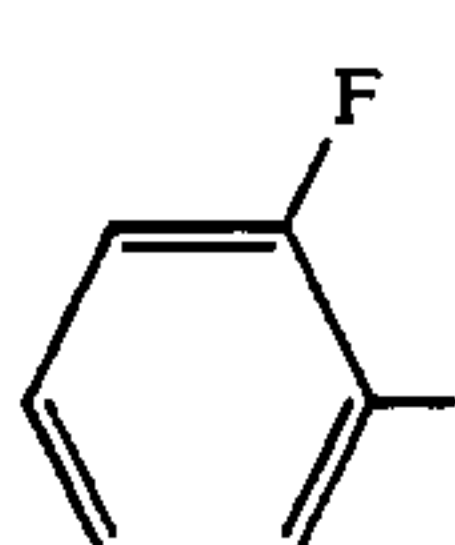
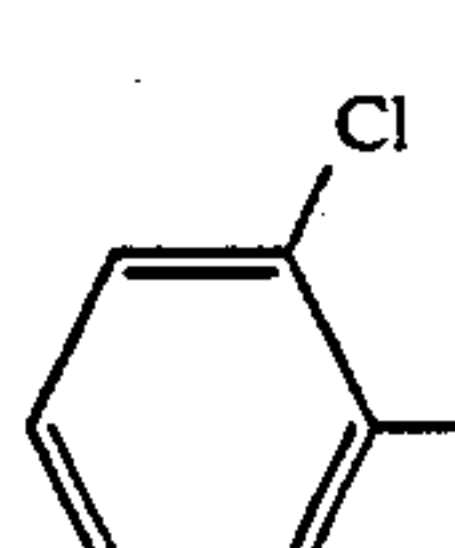
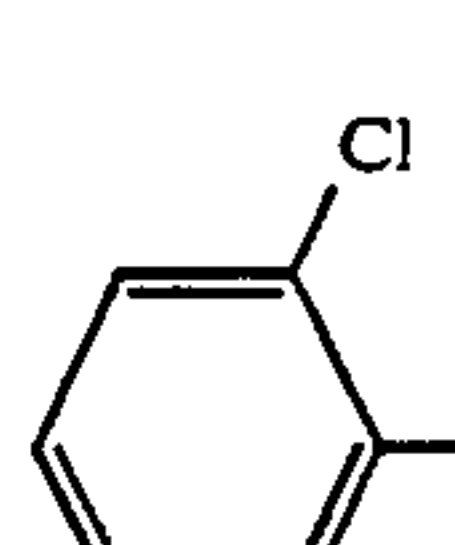
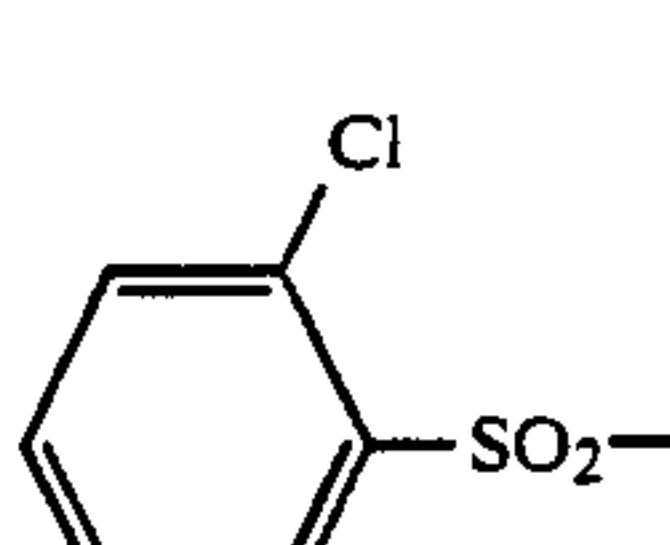
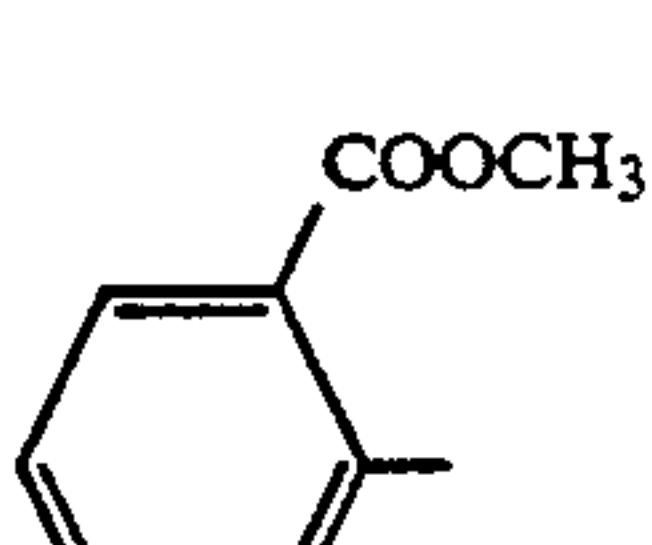
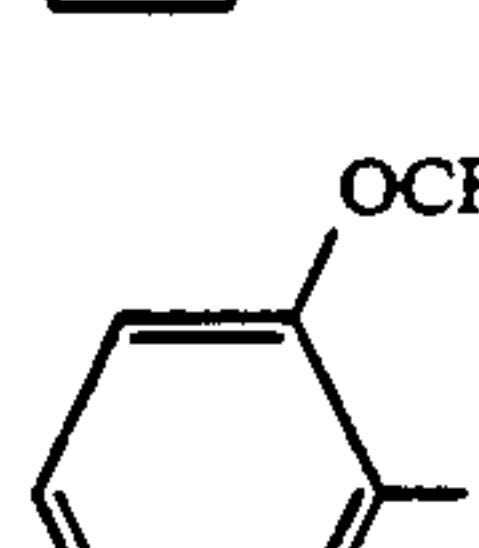
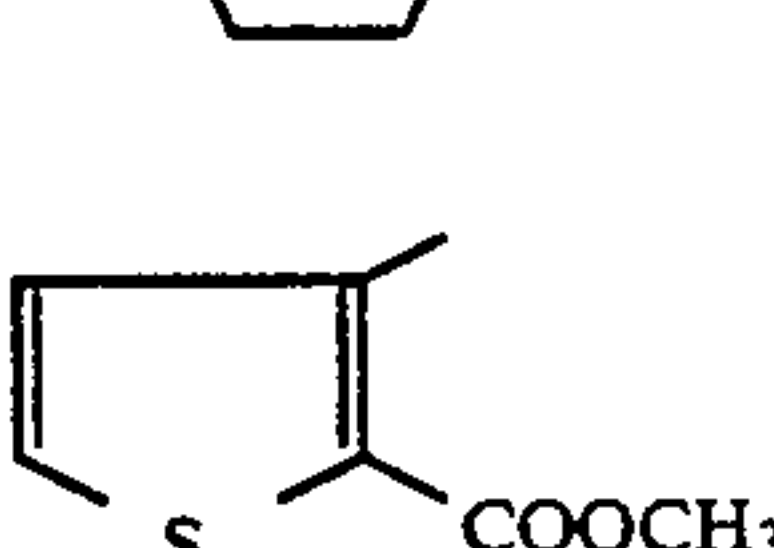
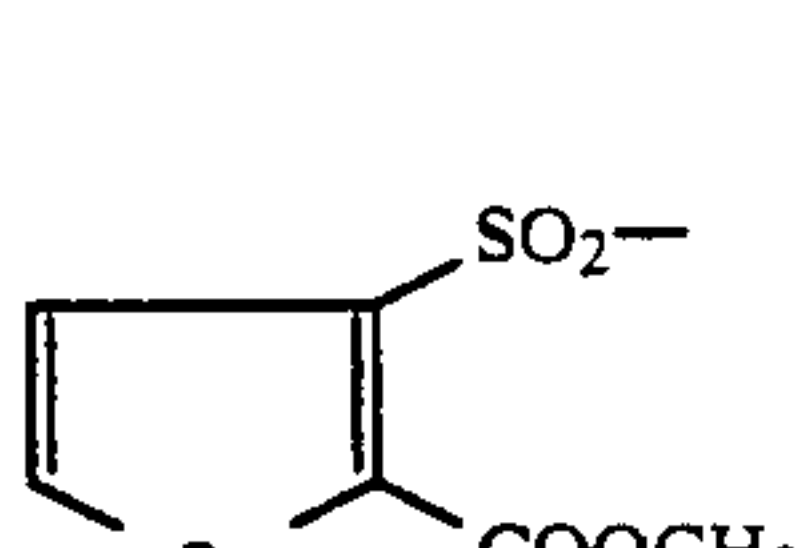
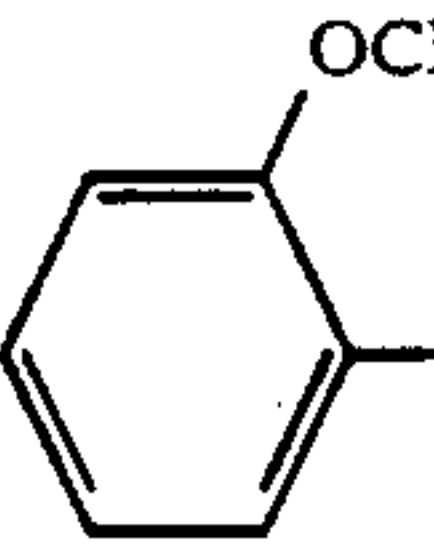
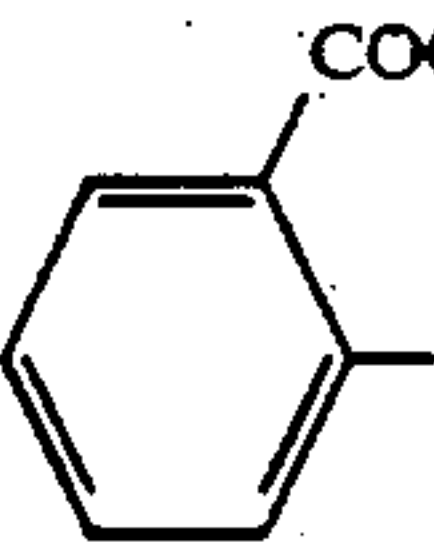
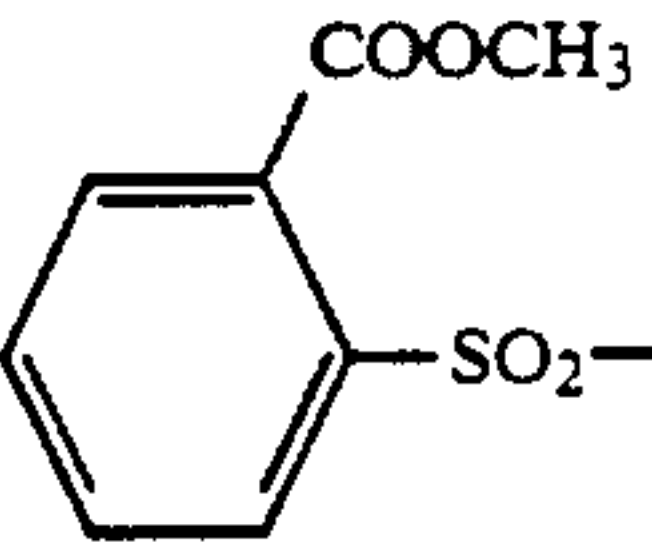
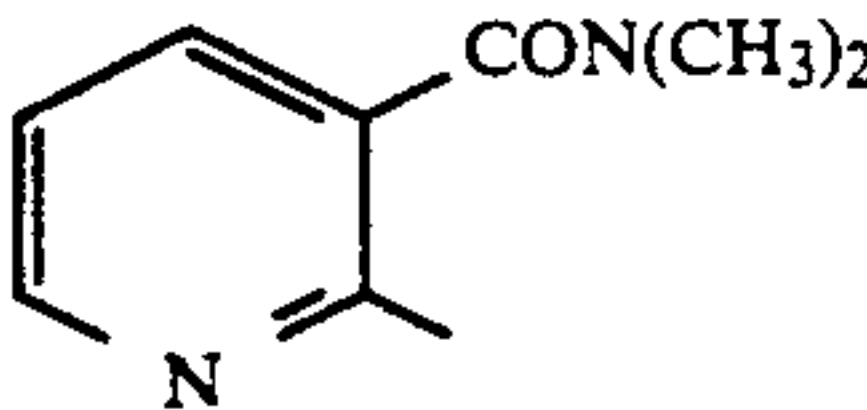
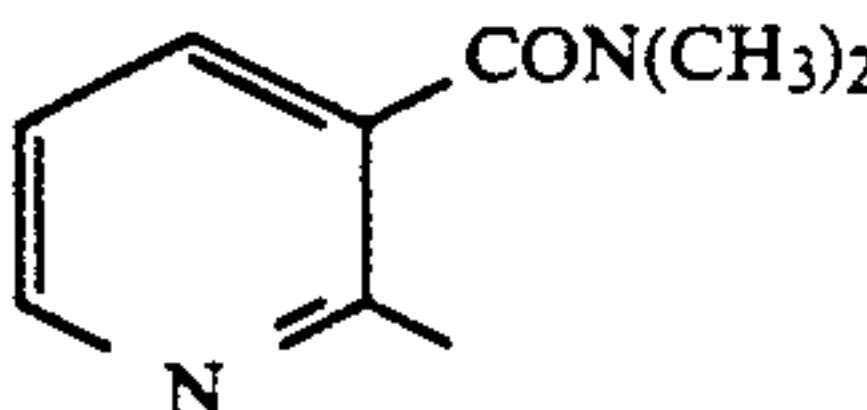
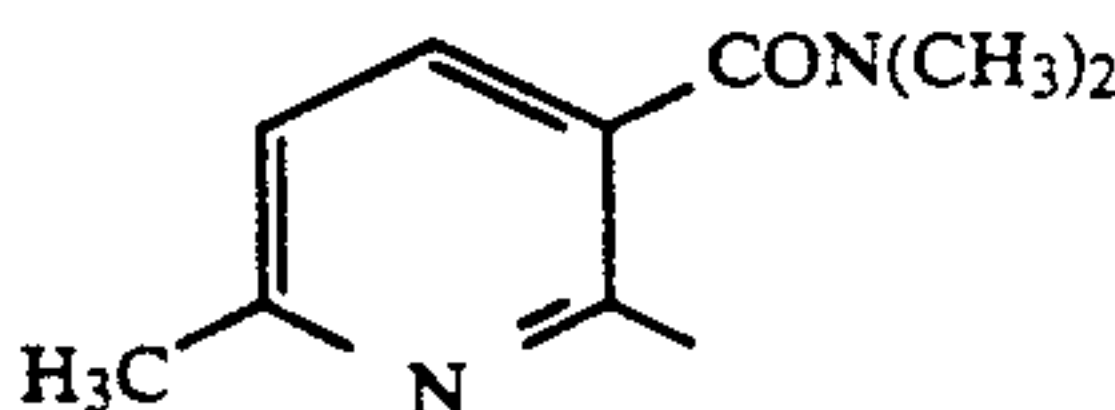
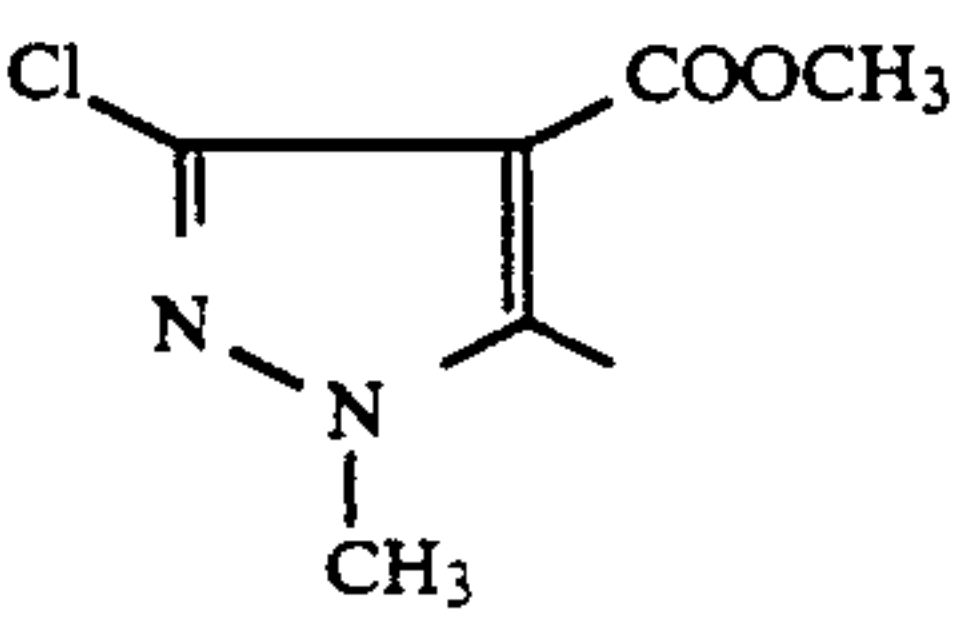
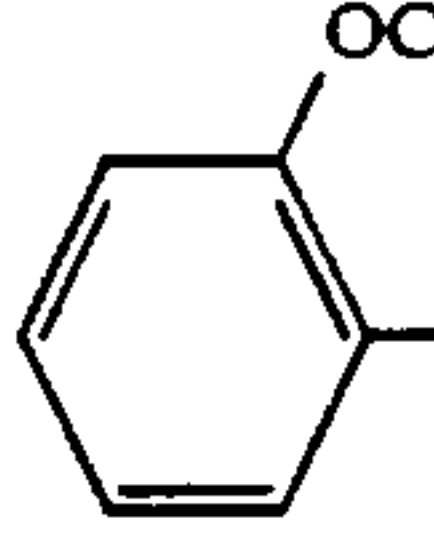
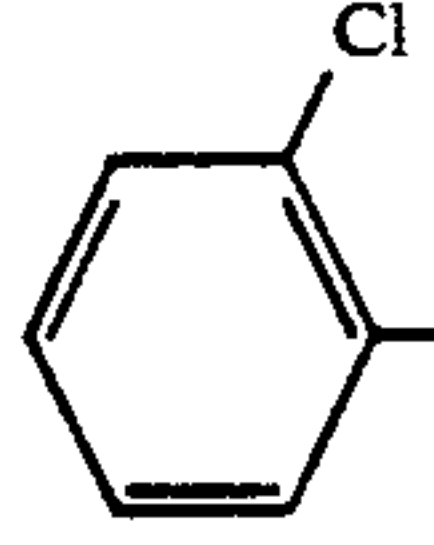
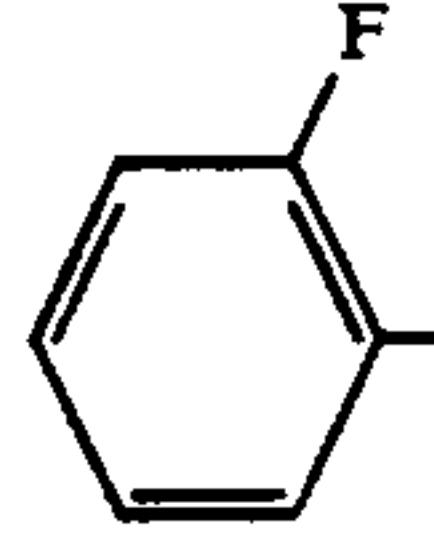
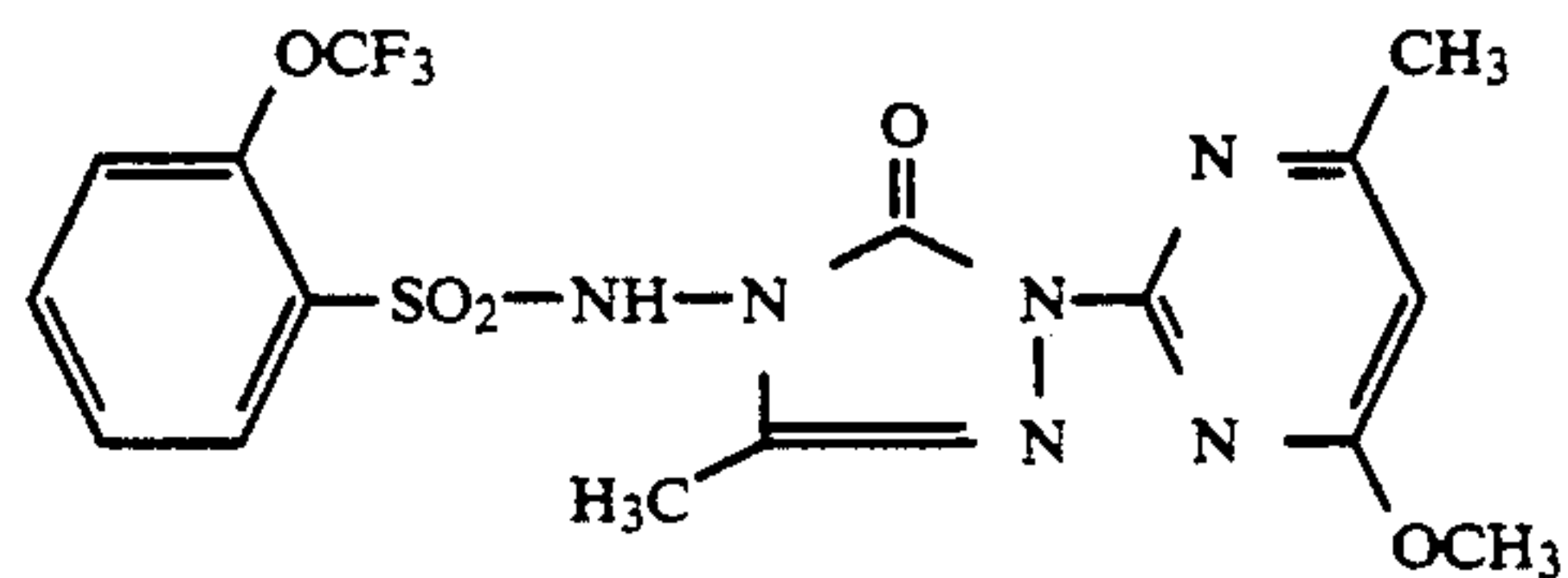
Ex- am- ple No.	Examples of the compounds of the formula (I)							Melting point (°C.)
	R ¹	R ²	R ³	R ⁴	X	Y	Z	
213			CH ₃	CH ₃	N	CH	C—OCH ₃	
214			CH ₃	CH ₃	N	CH	C—OCH ₃	
215			CH ₃	CH ₃	N	CH	C—OCH ₃	
216		H	CH ₃	CH ₃	N	CH	C—OCH ₃	
217		H	CH ₃	CH ₃	N	CH	C—OCH ₃	232
218		H	CH ₃	CH ₃	N	CH	C—OCH ₃	247
219			CH ₃	CH ₃	N	CH	C—OCH ₃	203
220		H	CH ₃	CH ₃	N	CH	C—OCH ₃	
221		H	CH ₃	CH ₃	N	CH	C—OCH ₃	248
222			CH ₃	CH ₃	N	CH	C—CH ₃	206

TABLE 2-continued

Ex- am- ple No.	Examples of the compounds of the formula (I)							Melting point (°C.)
	R ¹	R ²	R ³	R ⁴	X	Y	Z	
223		H	CH ₃	CH ₃	N	CH	C-CH ₃	308
224			CH ₃	CH ₃	N	CH	C-CH ₃	195
225		H	H	OCH ₃	N	CH	C-OCH ₃	
226		H	CH ₃	OCH ₃	N	CH	C-OCH ₃	
227		H	CH ₃	OCH ₃	N	CH	C-OCH ₃	
228		H	SCH ₃	OCH ₃	N	CH	C-OCH ₃	
229		H	CH ₃	OCH ₃	N	N	C-OCH ₃	179
230		H	CH ₃	OCH ₃	N	N	C-OCH ₃	(amor- phous)
231		H	CH ₃	OCH ₃	N	N	C-OCH ₃	87

60



The preparation in accordance with process (b) of the compound listed as Example No. 221 in Table 2 is described below by way of example: 65

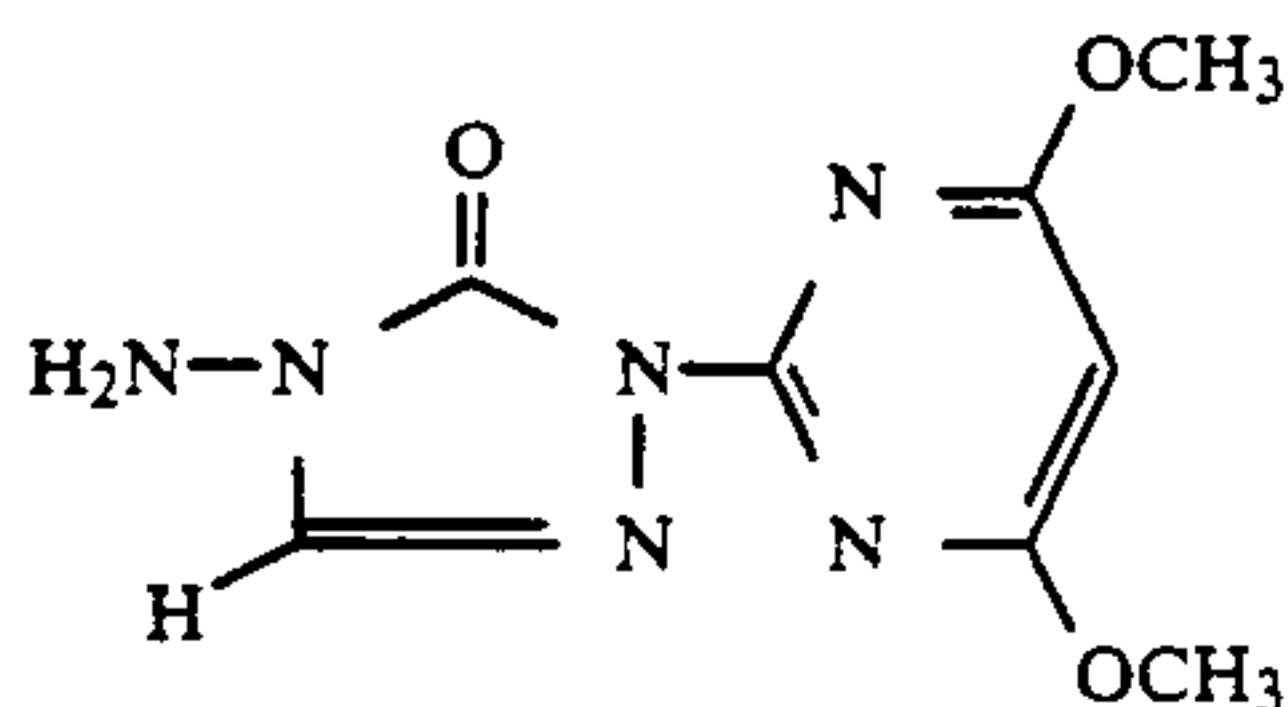
(process (b))

A mixture of 23.1 g (0.070 mol) of 4-(2-trifluoromethoxyphenylsulphonylamino)-5-methyl-2,4-dihydro-3H-1,2,4-triazol-3-one, 14.5 g (0.072 mol) of 4-methoxy-6-methyl-2-methylsulphonyl-pyrimidine, 50 g of potassium carbonate and 300 ml dioxane is refluxed for 6 hours. The mixture is allowed to cool and then filtered, the filter residue is washed with water and then taken up in methylene chloride and 5% strength hydrochloric acid, and the mixture is shaken. The organic phase is dried using sodium sulphate and filtered. After the filtrate has been concentrated, the residue is made to crystallize by trituration with ethanol, and the crystalline product is isolated by filtering off with suction.

6.0 g (21% of theory) of 4-(2-trifluoromethoxyphenylsulphonylamino)-5-methyl-2-(4-methoxy-6-methylpyrimidin-2-yl)-2,4-dihydro-3H-1,2,4-triazol-3-one of melting point 248° C. are obtained.

Starting Substances of the Formula (II)

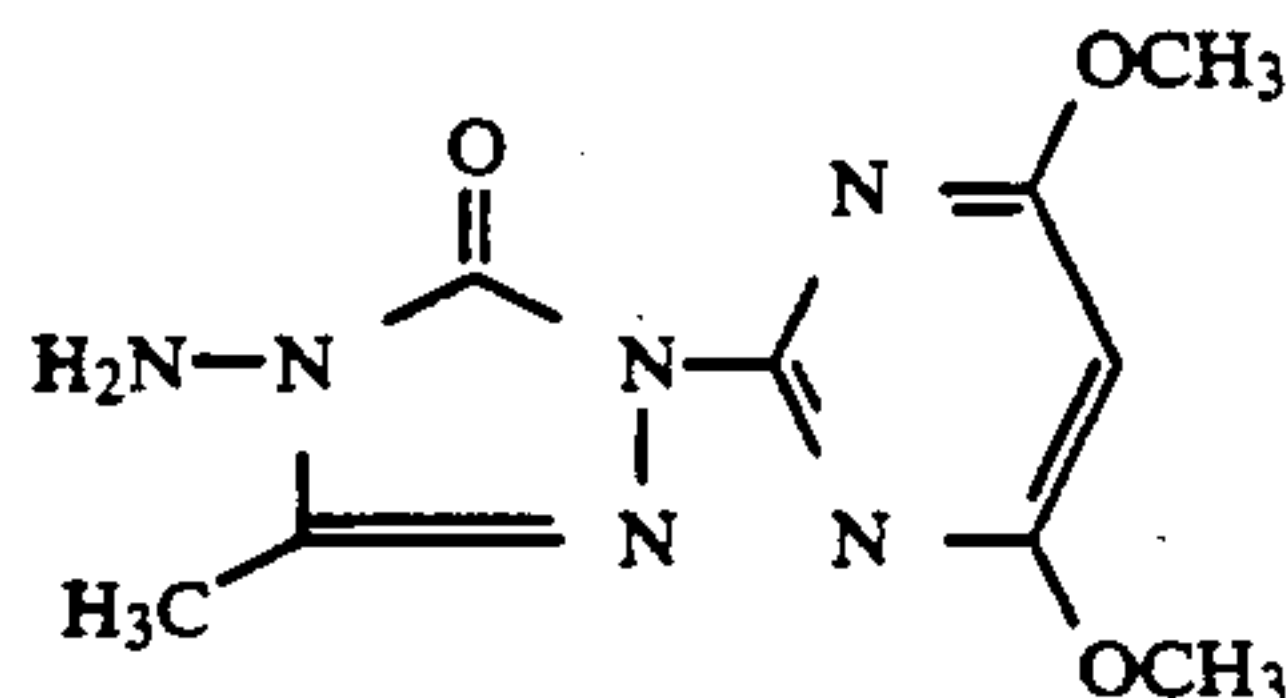
Example (II-1)



A mixture of 7.0 g (0.05 mol) of 4-(1-methyl-2,4-dihydro-3H-1,2,4-triazol-3-one), 21 g (0.15 mol) of potassium carbonate, 11.0 g (0.05 mol) of 4,6-dimethoxy-2-methylsulphonyl-pyrimidine and 100 ml of acetonitrile is refluxed for 3 hours. After the mixture has cooled, it is filtered and the filtrate is concentrated, the residue is taken up in 100 ml of ethanol/water (vol. 1:1), and the mixture is stirred for 3 hours at 60° C. after 1 ml of concentrated hydrochloric acid has been added. The mixture is concentrated, the residue is then triturated with ethanol, and the product obtained in the form of crystals is isolated by filtering off with suction.

9.7 g (82% of theory) of 4-amino-2-(4,6-dimethoxy-pyrimidin-2-yl)-2,4-dihydro-3H-1,2,4-triazol-3-one of melting point 238° C. are obtained.

Example (II-2)

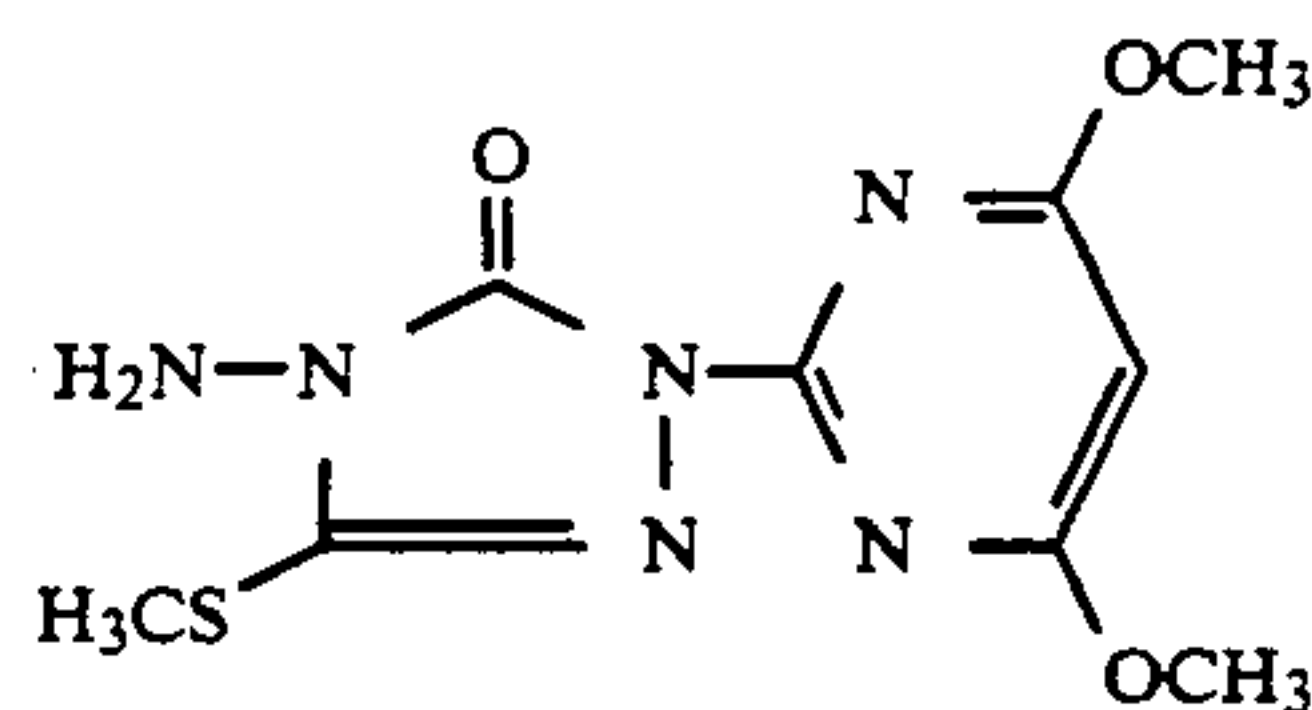


A mixture of 4.0 g (0.02 mol) of 4-(1,3-dimethylbutylideneamino)-5-methyl-2,4-dihydro-3H-1,2,4-triazol-3-one, 8.3 g (0.06 mol) of potassium carbonate, 4.4 g (0.02 mol) of 4,6-dimethoxy-2-methylsulphonyl-pyrimidine and 100 ml of acetonitrile is refluxed for 3 hours. After the mixture has cooled it is filtered, the filtrate is concentrated, the residue is taken up in 100 ml

of ethanol/water (vol. 1:1) and stirred for 60 minutes at 60° C. after 2 drops of concentrated hydrochloric acid have been added. The mixture is concentrated, the residue is stirred with saturated sodium hydrogen carbonate solution, and the crystalline product is isolated by filtering off with suction.

2.6 g (52% of theory) of 4-amino-5-methyl-2-(4,6-dimethoxy-pyrimidin-2-yl)-2,4-dihydro-3H-1,2,4-triazol-3-one of melting point 219° C. are obtained.

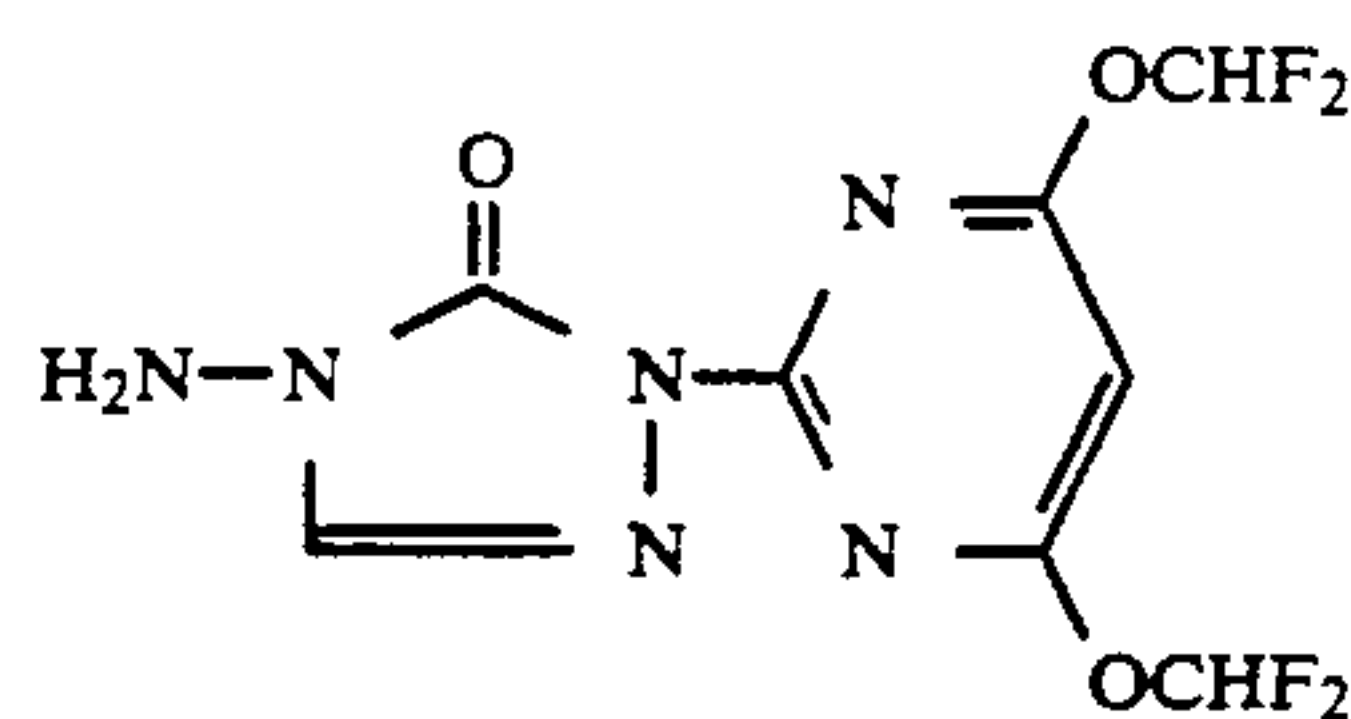
Example (II-3)



A mixture of 3.0 g (0.02 mol) of 4-amino-5-methylthio-2,4-dihydro-3H-1,2,4-triazol-3-one and 200 ml of acetone is refluxed for 4 hours after 0.1 g of p-toluenesulphonic acid has been added. The mixture is concentrated, the residue is taken up in 100 ml of acetonitrile, and the reaction mixture is refluxed for 12 hours after 8.3 g (0.06 mol) of potassium carbonate and 4.4 g (0.02 mol) of 4,6-dimethoxy-2-methylsulphonyl-pyrimidine have been added. The cooled mixture is filtered, the filtrate is concentrated, the residue is taken up in 100 ml of ethanol/water (vol. 1:1) and stirred for 3 hours at 60° C. after 2 ml of concentrated hydrochloric acid have been added. The mixture is concentrated, the residue is stirred with saturated sodium hydrogen carbonate solution, and the crystalline product is isolated by filtering off with suction.

4.9 g (86% of theory) of 4-amino-5-methylthio-2-(4,6-dimethoxy-pyrimidin-2-yl)-2,4-dihydro-3H-1,2,4-triazol-3-one of melting point 198° C. are obtained.

Example (II-4)

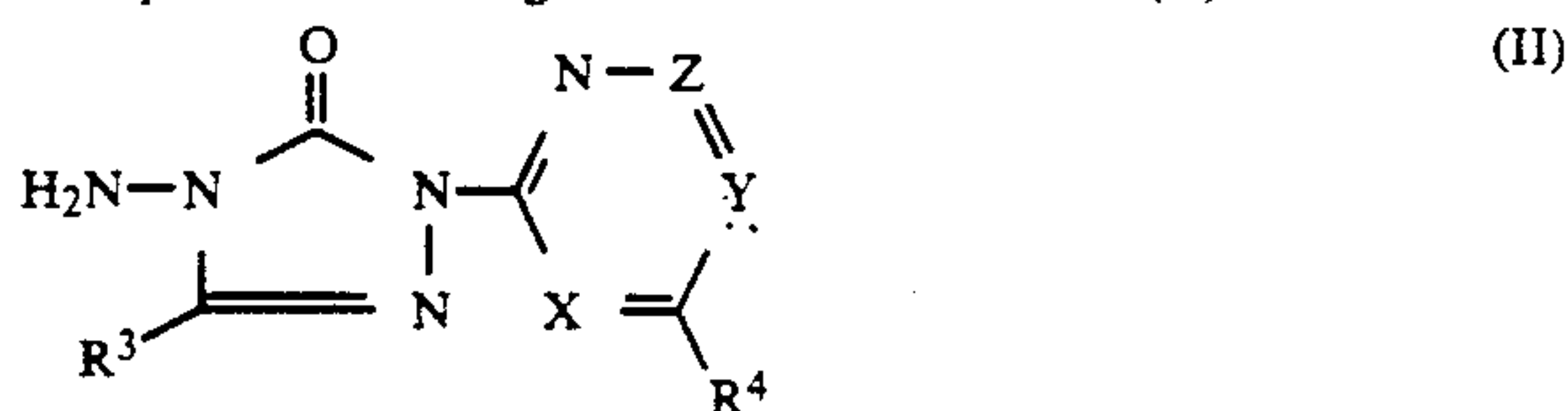


A mixture of 3.0 g (0.03 mol) of 4-amino-2,4-dihydro-3H-1,2,4-triazol-3-one, 8.7 g (0.03 mol) of 2-methanesulphonyl-4,6-bis-difluoromethoxy-pyrimidine and 12.4 g (0.09 mol) of potassium carbonate in 100 ml of dioxane is heated at 60° C. for approximately 4 hours with stirring. The mixture is concentrated in vacuo, the residue is stirred with water, and the insoluble precipitate is filtered off with suction and washed to neutrality using highly dilute hydrochloric acid and water. After drying, 7.0 g (0.0226 mol = 75% of theory) of 4-amino-2-(4,6-bis-difluoromethoxy)pyrimidin-2-yl-2,4-dihydro-3H-1,2,4-triazol-3-one of melting point 195° C. remain.

The compounds of the formula (II) which are listed in Table 3 below can be prepared analogously to Examples (II-1) to (II-4).

TABLE 3

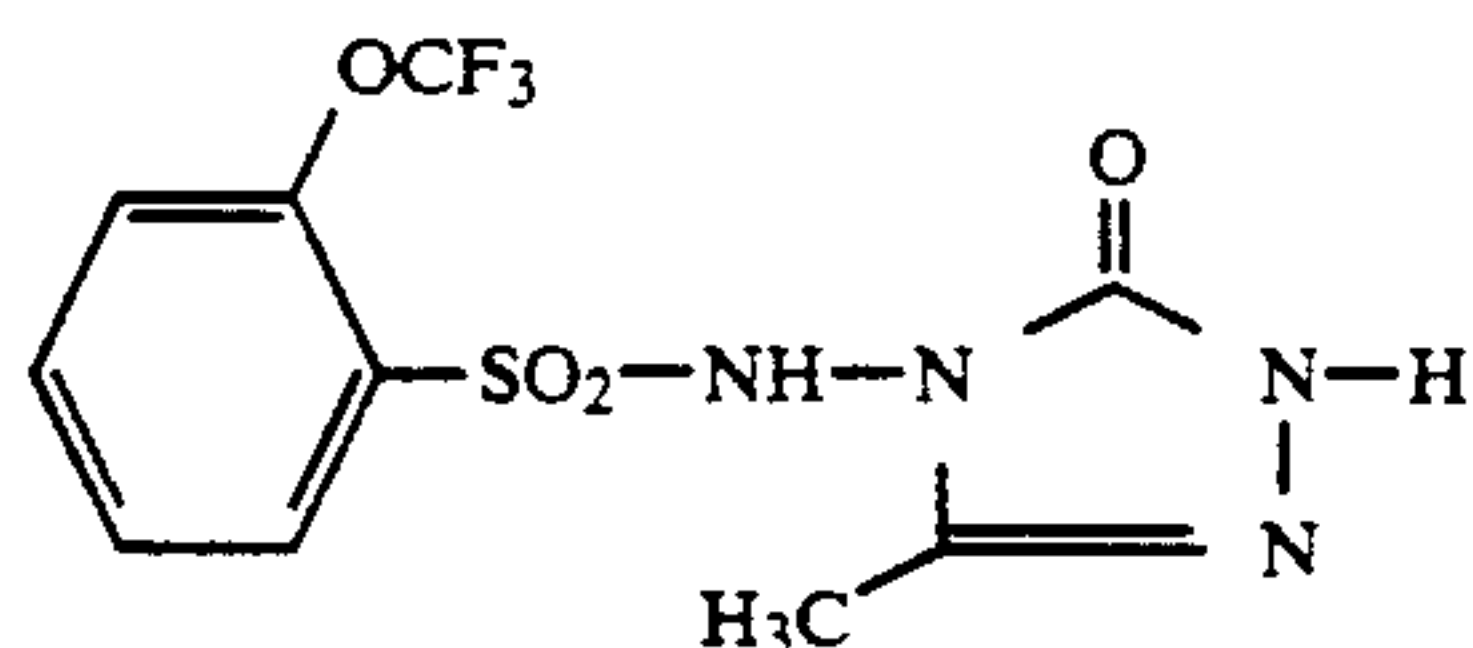
Preparation examples of the starting substances of the formula (II)



Ex. No.	R ³	R ⁴	X	Y	Z	Melting point (°C.)
II-5	C(CH ₃) ₃	OCH ₃	N	CH	C-OCH ₃	205
II-6	CF ₃	OCH ₃	N	CH	C-OCH ₃	213
II-7	C ₂ H ₅	OCH ₃	N	CH	C-OCH ₃	167
II-8	C ₃ H ₇	OCH ₃	N	CH	C-OCH ₃	183
II-9	CH(CH ₃) ₂	OCH ₃	N	CH	C-OCH ₃	206
II-10	CH ₃	CH ₃	N	CH	C-CH ₃	310
II-11	H	OCH ₃	N	CH	C-NH-CH ₃	
II-12	H	OCH ₃	N	N	C-OCH ₃	
II-13	CH ₃	OCH ₃	N	N	C-CH ₃	
II-14	CH ₃	Cl	N	CH	C-OCH ₃	
II-15		OCH ₃	N	CH	C-OCH ₃	
II-16		OCH ₃	N	CH	C-OCH ₃	
II-17	CH ₃	OCHF ₂	N	CH	C-OCHF ₂	255

Starting Substances of the Formula (IV)

Example (IV-1)



10.0 g (0.088 mol) of 4-amino-5-methyl-2,4-dihydro-3H-1,2,4-triazol-3-one are dissolved in 50 ml of dry pyridine, the mixture is cooled to -10° C., and 32.7 g (0.12 mol) of 2-trifluoromethoxy-benzenesulphonyl chloride are added. The reaction mixture is allowed to

reach room temperature (approximately 20° C.) and stirred for approximately 12 hours. After the mixture has been concentrated, the residue is taken up in methylene chloride and shaken with 1N hydrochloric acid and then with water. The organic phase is dried with sodium sulfate and filtered. The solvent is carefully distilled off from the filtrate under a water pump vacuum. 23.1 g (80% of theory) of 4-(2-trifluoromethoxyphenylsulphonylamino)-5-methyl-2,4-dihydro-3H-1,2,4-triazol-3-one are obtained as an oily residue.

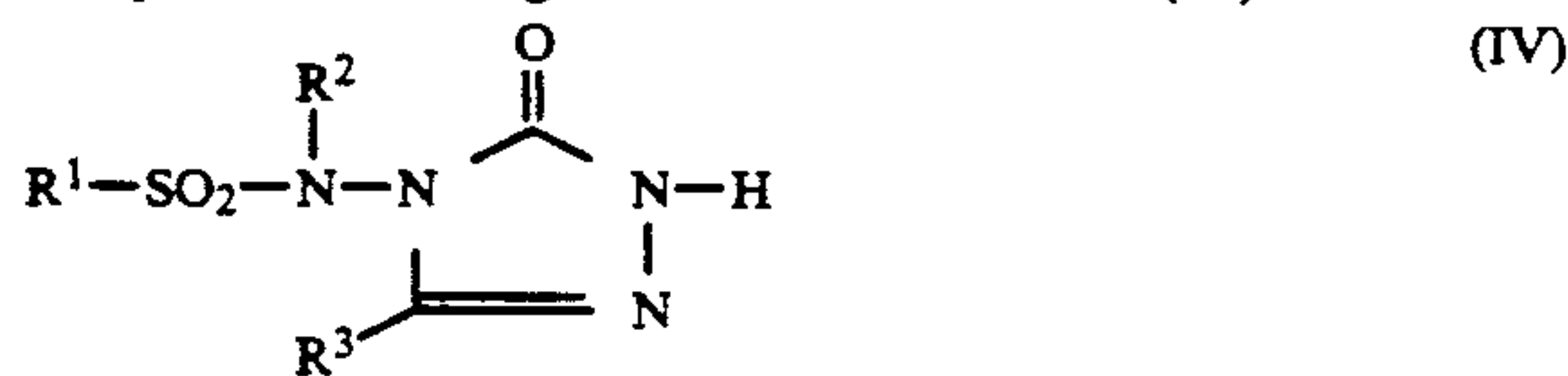
¹H-NMR*) (DMSO, δ, ppm): 2.10 (s, CH₃), 7.55, 7.83, 7.93 (m, 4H), approximately 11.5 (m, 2×NH).

*) The ¹H-NMR Spectra were recorded in dimethyl sulphoxide (DMSO) using tetramethylsilane (TMS) as the internal standard. The chemical shift is indicated as the -value in ppm.

The compounds of the formula (IV), which are listed in Table 4 below, can be prepared analogously.

TABLE 4

Preparation Examples of the starting substances of the formula (IV)



Example No.	R ¹	R ²	R ³	Physical data
IV-2	COOCH ₃	H	CH ₃	NMR: 2.04 ^{a)}

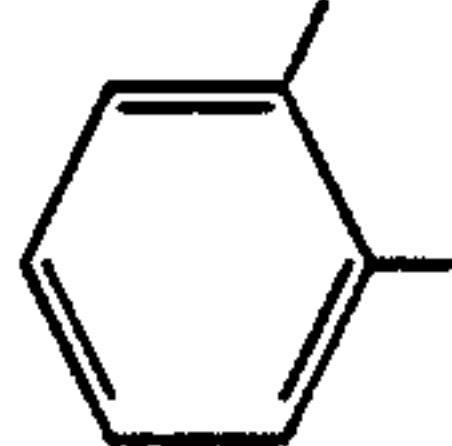
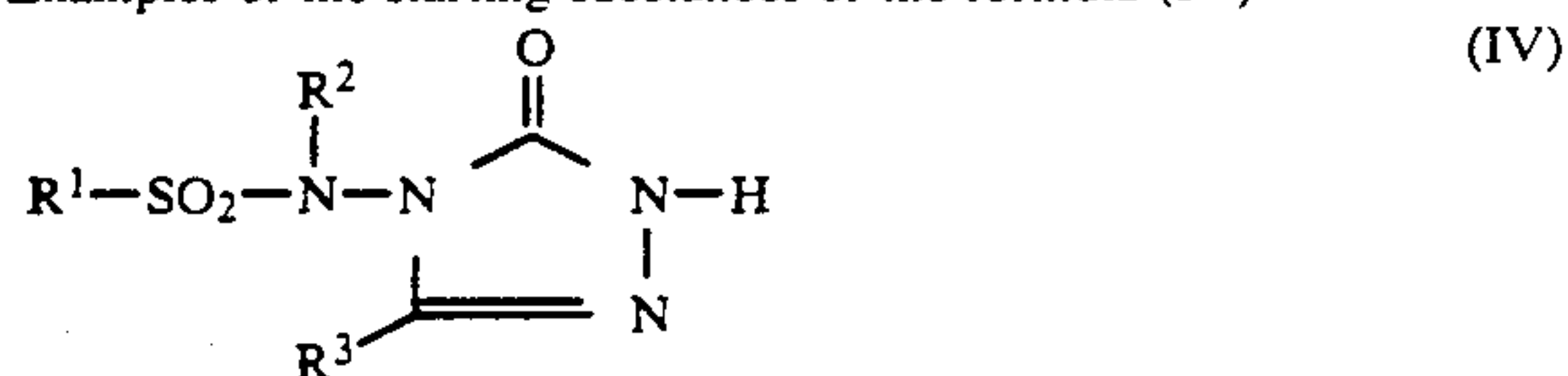


TABLE 4-continued

Preparation Examples of the starting substances of the formula (IV)

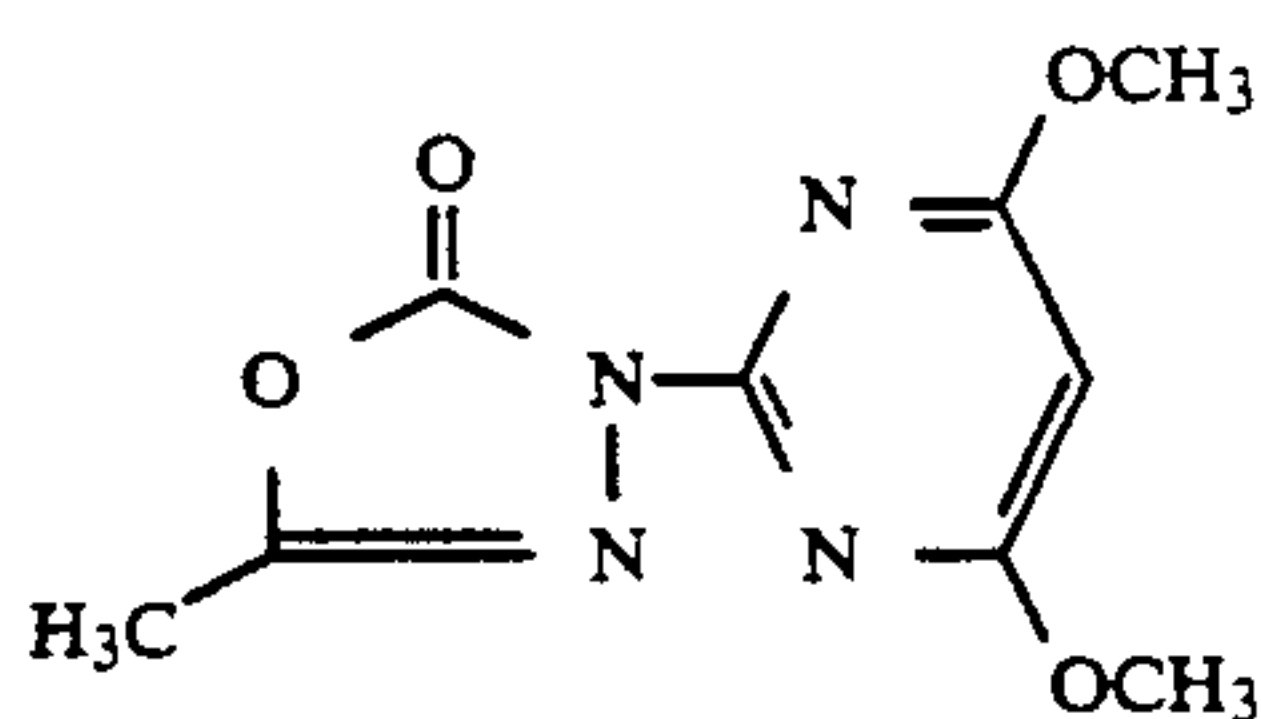


Example No.	R ¹	R ²	R ³	Physical data
IV-3		H	CH ₃	NMR: 2.04 ^{a)}
IV-4		H	CH ₃	NMR: 2.11 ^{a)}
IV-5			CH ₃	M.p.: 180-190° C.
IV-6		H	CH ₃	M.p.: 206-207° C.

^{a)}H-NMR (DMSO, δ, ppm) for R³ = CH₃

Starting Substances of the Formula (XI)

Example (XI-1)

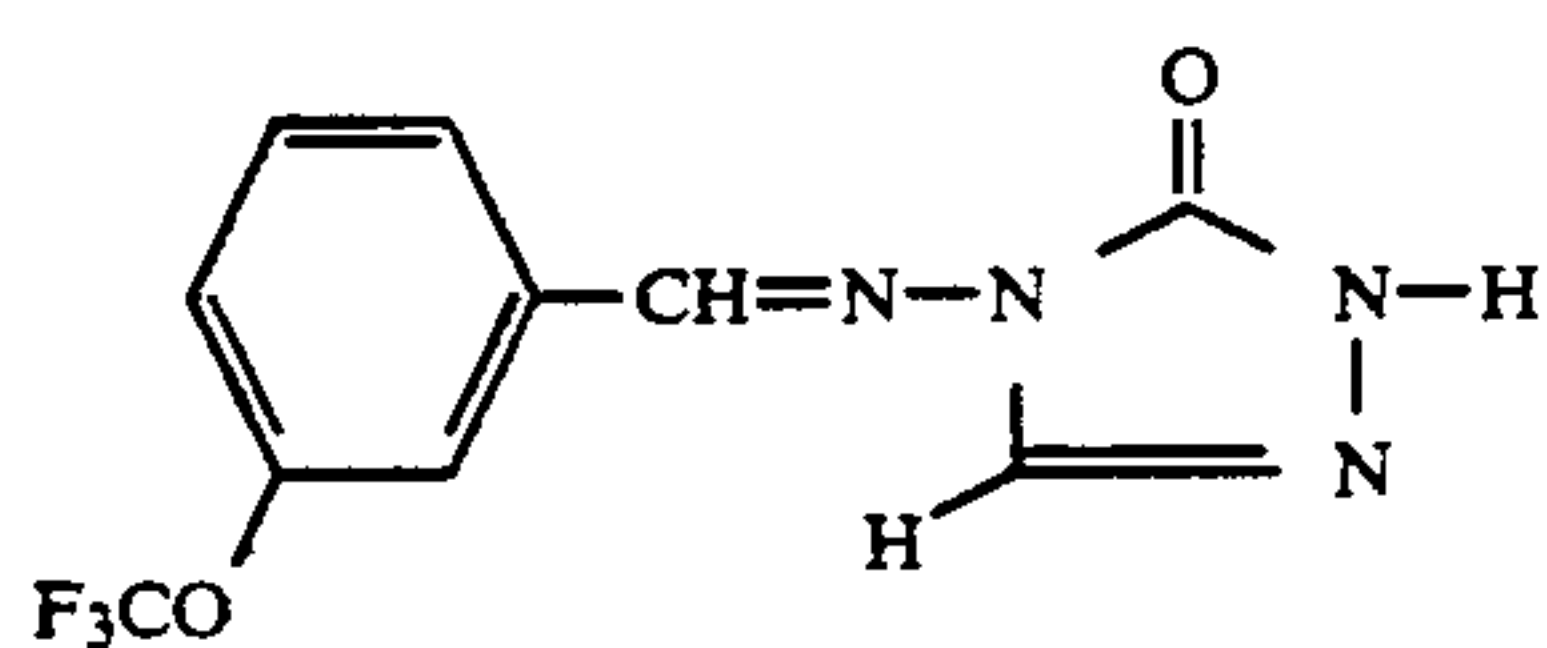


A mixture of 4.0 g (0.04 mol) of 5-methyl-1,3,4-oxadiazol-2(3H)-one, 8.8 g (0.047 mol) of 4,6-dimethoxy-2-methylsulphonyl-pyrimidine, 16.9 g (0.12 mol) of potassium carbonate and 100 ml of acetonitrile is stirred for 4 hours at 60° C. The cold mixture is concentrated under a water-pump vacuum, the residue is stirred with water, and the crystalline product is isolated by filtering off with suction.

4.8 g (50% of theory) of 3-(4,6-dimethoxy-2-methyl-1,3,4-oxadiazol-2(3H)-one)-5-methyl-1,3,4-oxadiazol-2(3H)-one of melting point 158° C. are obtained.

USE EXAMPLES

In the following Use Examples, the compound listed below is used as comparison substance:



35 4-(3-trifluoromethoxy-benzylideneamino)-2,4-dihydro-3H-1,2,4-triazol-3-one
(disclosed in U.S. Pat. No. 3,884,910).

Example A

40 Post-emergence test
Solvent: 5 parts by weight of acetone
Emulsifier: 1 part by weight of alkylaryl polyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amount of solvent, the stated amount of emulsifier is added and the concentrate is diluted with water to the desired concentration.

Test plants which have a height of 5-15 cm are sprayed with the preparation of the active compound in such a way as to apply the particular amounts of active compound desired per unit area. The concentration of the spray liquor is chosen such that the particular amounts of active compound desired are applied in 2,000 l of water/ha. After three weeks, the degree of damage to the plants is rated in % damage in comparison to the development of the untreated control.

The figures denote:

0% = no action (like untreated control)

60 100% = total destruction

(A) In this test, for example, the compounds according to Preparation Examples: (1), (2), (3), (4), (92), (208) and (210) show a clearly superior activity compared with the prior art.

65

Example B

Pre-emergence test

Solvent: 5 parts by weight of acetone

Emulsifier: 1 part by weight of alkylaryl polyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amount of solvent, the stated amount of emulsifier is added and the concentrate is diluted with water to the desired concentration.

Seeds of the test plants are sown in normal soil and, after 24 hours, watered with the preparation of the active compound. It is expedient to keep constant the amount of water per unit area. The concentration of the active compound in the preparation is of no importance, only the amount of active compound applied per unit area being decisive. After three weeks, the degree of damage to the plants is scored in % damage in comparison to the development of the untreated control. The figures denote:

0% = no action (like untreated control)

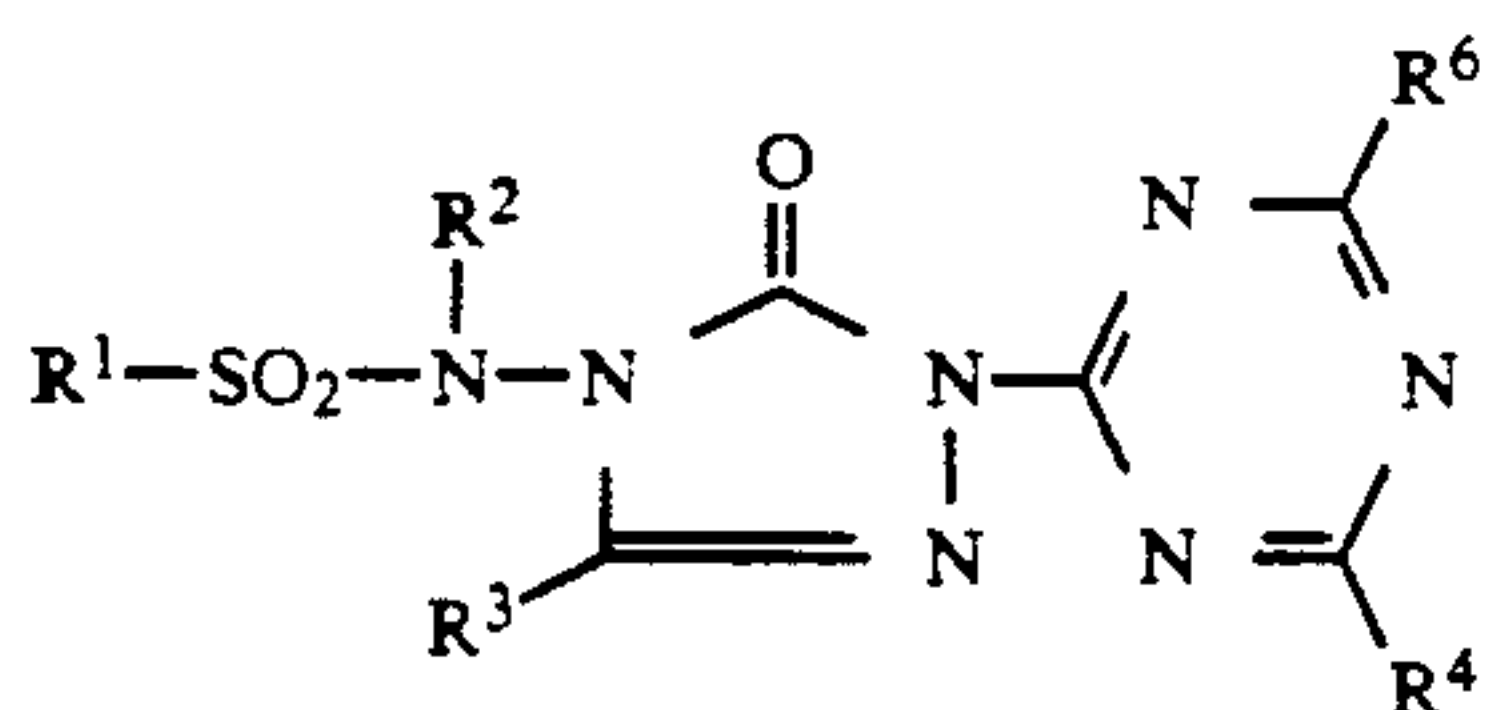
100% = total destruction

In this test, for example the compounds of Preparation Examples: (1), (2), (3), (4), (208) and (210) have a clearly superior activity compared with the prior art.

It will be appreciated that the instant specification and claims are set forth by way of illustration and not limitation, and that various modifications and changes may be made without departing from the spirit and scope of the present invention.

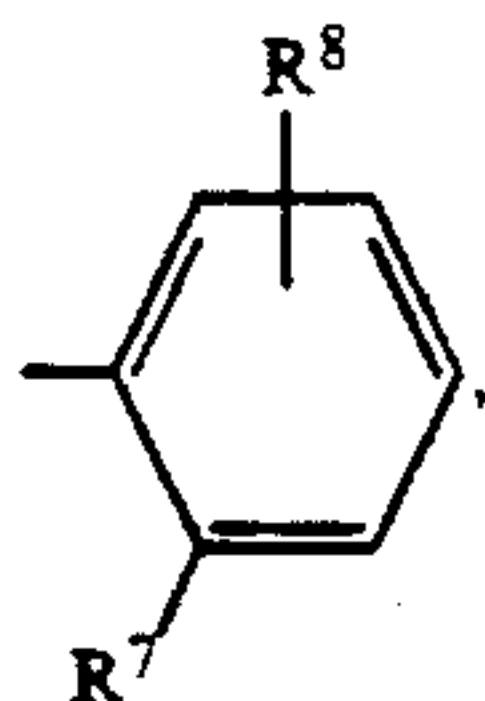
What is claimed is:

1. A compound of the formula



in which

R¹ represents the radical



where

R⁷ and R⁸ are identical or different and represent hydrogen, fluorine, chlorine, bromine, iodine, cyano, nitro, C₁-C₆-alkoxy (which is optionally substituted by fluorine, chlorine, bromine, cyano, carboxyl, C₁-C₄-alkoxycarbonyl, C₁-C₄-alkylaminocarbonyl, di-(C₁-C₄-alkyl)aminocarbonyl, hydroxyl, C₁-C₄-alkoxy, formyloxy, C₁-C₄-alkyl-carbonyloxy, C₁-C₄-alkoxycarbonyloxy, C₁-C₄-alkylamino-carbonyloxy, C₁-C₄-alkylthio, C₁-C₄-alkylsulphinyl, C₁-C₄-alkylsulphonyl, di-((C₁-C₄-alkyl)aminosulphonyl, C₃-C₆-cycloalkyl or phenyl), represent C₂-C₆-alkenyl (which is optionally substituted by fluorine, chlorine, bromine, cyano, C₁-C₄-alkoxycarbonyl, carboxyl or phenyl), represent C₂-C₆-alkinyl (which is optionally substituted by fluorine, chlorine, bromine, cyano, C₁-C₄-alkoxy-carbonyl, carboxyl or phenyl), represent C₁-C₄-alkoxy (which is optionally substituted by

fluorine, chlorine, bromine, cyano, carboxy, C₁-C₄-alkoxy-carbonyl, C₁-C₄-alkoxy, C₁-C₄-alkylthio, C₁-C₄-alkylsulphinyl or C₁-C₄-alkylsulphonyl), represent C₁-C₄-alkylthio (which is optionally substituted by fluorine, chlorine, bromine, cyano, carboxyl, C₁-C₄-alkoxy-carbonyl, C₁-C₄-alkylthio, C₁-C₄-alkylsulphinyl or C₁-C₄-alkylsulphonyl), represent C₃-C₆-alkenyloxy (which is optionally substituted by fluorine, chlorine, bromine, cyano or C₁-C₄-cyano or C₁-C₄-alkoxy-carbonyl), represent C₂-C₆-alkenylthio (which is optionally substituted by fluorine, chlorine, bromine, cyano, nitro, C₁-C₃-alkylthio or C₁-C₄-alkoxycarbonyl), C₃-C₆-alkinyloxy, C₃-C₆-alkinylthio or represent the radical —S(O)_p—R⁹, where p represents the numbers 1 or 2 and

R⁹ represents fluorine, C₁-C₄-alkyl (which is optionally substituted by fluorine, chlorine, bromine, cyano or C₁-C₄-alkoxy-carbonyl), C₃-C₆-alkenyl, C₃-C₆-alkinyl, C₁-C₄-alkoxy, C₁-C₄-alkoxy-C₁-C₄-alkylamino, C₁-C₄-alkylamino, di-(C₁-C₄-alkyl)-amino or represents the radical —NOR¹⁰, where

R¹⁰ represents C₁-C₆-alkyl (which is optionally substituted by fluorine, chlorine, cyano, C₁-C₄-alkoxy, C₁-C₄-alkylthio, C₁-C₄-alkylsulphinyl, C₁-C₄-alkylsulphonyl, C₁-C₄-alkyl-carbonyl, C₁-C₄-alkoxycarbonyl, C₁-C₄-alkylaminocarbonyl or di-(C₁-C₄-alkyl)amino-carbonyl), represents C₃-C₆-alkenyl (which is optionally substituted by fluorine, chlorine or bromine), C₃-C₆-alkinyl, C₃-C₆-cycloalkyl, C₃-C₆-cycloalkyl-C₁-C₂-alkyl, phenyl-(C₁-C₂-alkyl (which is optionally substituted by fluorine, chlorine, nitro, cyano, C₁-C₄-alkyl, C₁-C₄-alkoxy or C₁-C₄-alkoxy-carbonyl), represents benzydryl or represents phenyl (which is optionally substituted by fluorine, chlorine, nitro, cyano, C₁-C₄-alkyl, trifluoromethyl, C₁-C₄-alkoxy, C₁-C₂-fluoroalkoxy, C₁-C₄-alkylthio, trifluoromethylthio or C₁-C₄-alkoxycarbonyl),

R⁷ and/or R⁸ furthermore represent phenyl or phenoxy, C₁-C₄-alkylcarbonylamino, C₁-C₄-alkoxy-carbonylamino, C₁-C₄-alkylaminocarbonyl-amino, di-(C₁-C₄-alkyl)-aminocarbonylamino, or represent the radical —CO—R¹¹, where

R¹¹ represents C₁-C₆-alkyl, C₁-C₆-alkoxy, C₃-C₆-cycloalkoxy, C₃-C₆-alkenyloxy, C₁-C₄-alkylthio, C₁-C₄-alkylamino, C₁-C₄-alkoxyamino, C₁-C₄-alkoxy-C₁-C₄-alkyl-amino or di-(C₁-C₄-alkyl)-amino (which are optionally substituted by fluorine and/or chlorine),

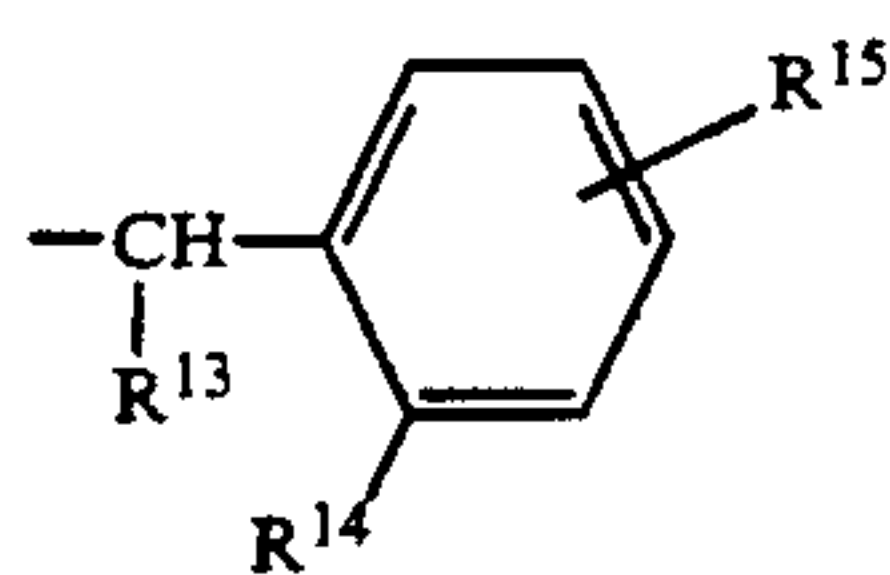
R⁷ and/or R⁸ furthermore represent C₁-C₄-alkylsulphonyloxy, di-(C₁-C₄-alkyl)-aminosulphonylamino or the radical —CH=N—R¹², where

R¹² represents C₁-C₆-alkyl which is optionally substituted by fluorine, chlorine, cyano, carboxyl, C₁-C₄-alkoxy, C₁-C₄-alkylthio, C₁-C₄-alkylsulphinyl or C₁-C₄-alkylsulphonyl, represents benzyl which is optionally substituted

by fluorine or chlorine, represents C₃-C₆-alkenyl or C₃-C₆-alkynyl, each of which is optionally substituted by fluorine or chlorine, represents phenyl which is optionally substituted by fluorine, chlorine, bromine, C₁-C₄-alkyl, C₁-C₄-alkoxy, trifluoromethyl, trifluoromethoxy or trifluoromethylthio, represents C₁-C₆-alkoxy, C₃-C₆-alkenoxy, C₃-C₆-alkinoxy or benzyloxy, each of which is optionally substituted by fluorine, and/or chlorine, represents amino, C₁-C₄-alkylamino, di-(C₁-C₄-alkyl)-amino, phenylamino, C₁-C₄-alkyl-carbonylamino, C₁-C₄-alkoxy-carbonylamino, C₁-C₄-alkylsulphonylamino or represents phenylsulphonylamino which is optionally substituted by fluorine, chlorine, bromine or methyl,

where furthermore

R¹ represents the radical

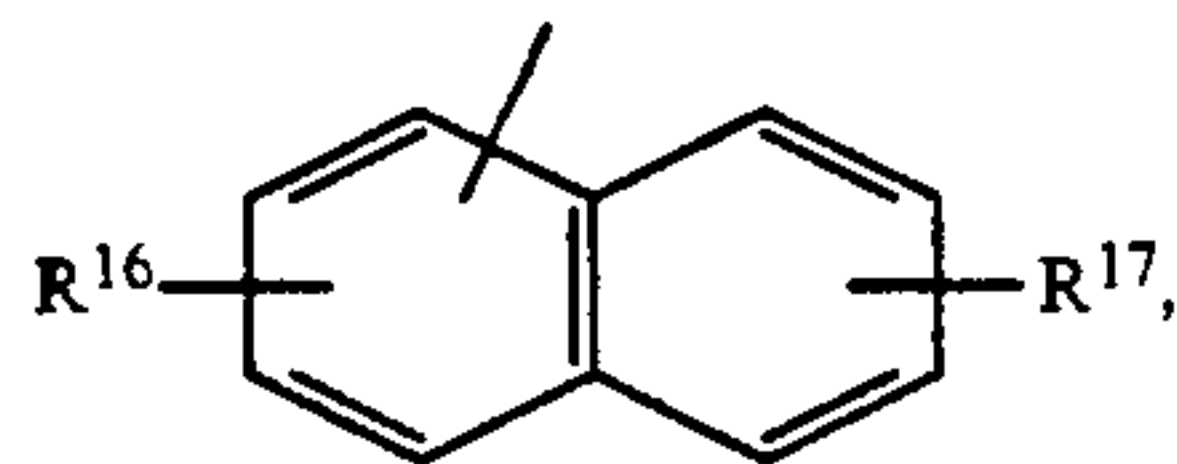


where

R¹³ represents hydrogen or C₁-C₄-alkyl,

R¹⁴ and R¹⁵ are identical or different and represent hydrogen, fluorine, chlorine, bromine, nitro, cyano, C₁-C₄-alkyl (which is optionally substituted by fluorine and/or chlorine), C₁-C₄-alkoxy (which is optionally substituted by fluorine and/or chlorine), carboxyl, C₁-C₄-alkoxycarbonyl, C₁-C₄-alkylsulphonyl or di-(C₁-C₄-alkyl)-aminosulphonyl; where furthermore

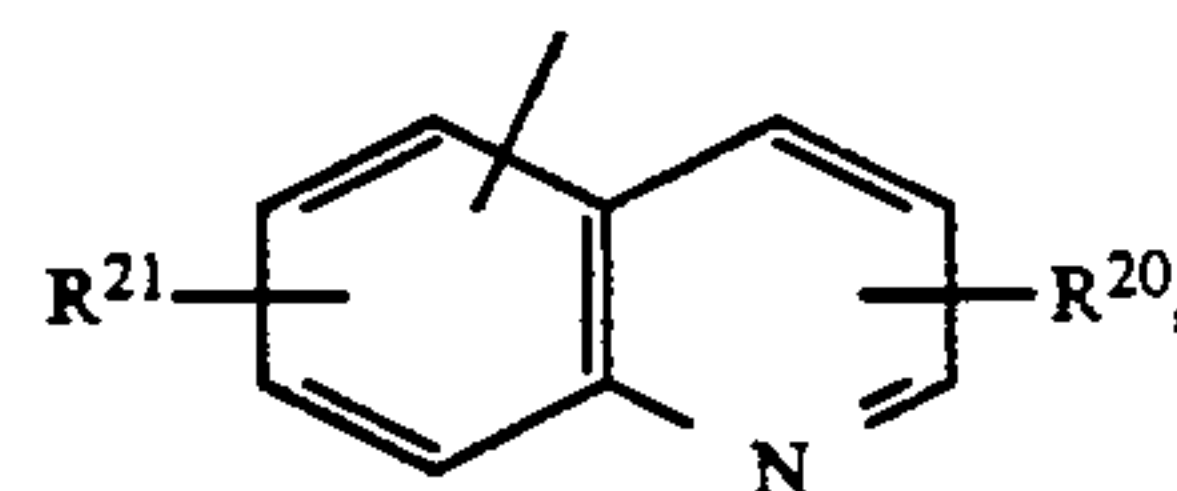
R¹ represents the radical



where

R¹⁶ and R¹⁷ are identical or different and represent hydrogen, fluorine, chlorine, bromine, nitro, cyano, C₁-C₄-alkyl (which is optionally substituted by fluorine and/or chlorine) or C₁-C₄-alkoxy (which is optionally substituted by fluorine and/or chlorine); where furthermore

R¹ represents the radical

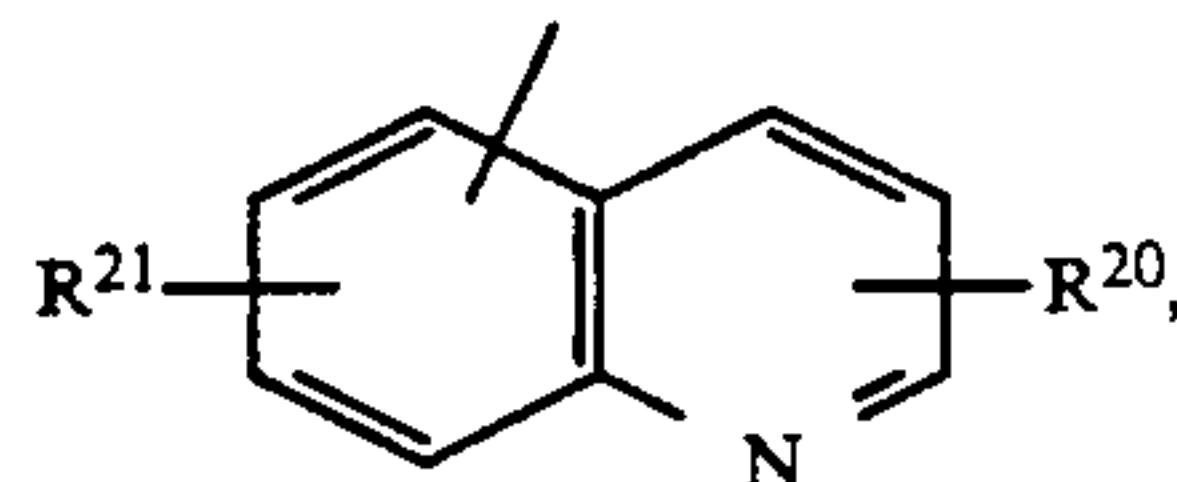


where

R¹⁸ and R¹⁹ are identical or different and represent hydrogen, fluorine, chlorine, bromine, nitro, cyano, C₁-C₄-alkyl (which is optionally substituted by fluorine and/or chlorine), C₂-C₄-alkenyl (which is optionally substituted by fluorine and/or chlorine), C₁-C₄-alkoxy (which is optionally substituted by fluorine and/or chlorine),

represents C₁-C₄-alkylthio, C₁-C₄-alkylsulphinyl or C₁-C₄-alkylsulphonyl (which are optionally substituted by fluorine and/or chlorine), and represents di-(C₁-C₄-alkyl)-aminosulphonyl, C₁-C₄-alkoxy-carbonyl, dimethylaminocarbonyl or dioxolanyl; where furthermore

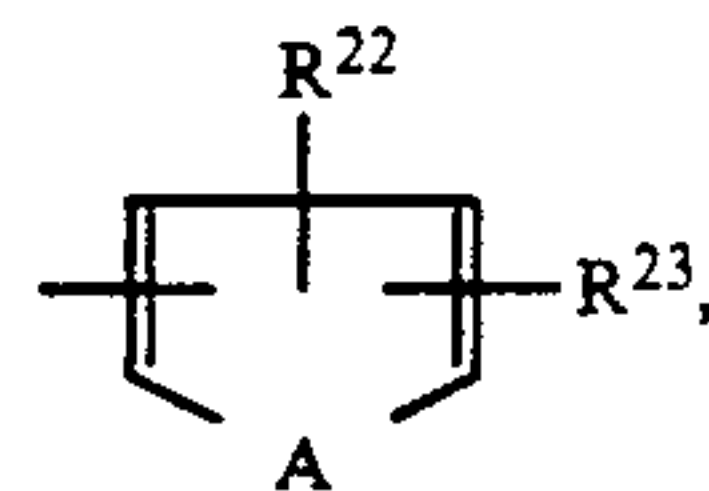
R¹ represents the radical



where

R²⁰ and R²¹ are identical or different and represent hydrogen, fluorine, chlorine, bromine, C₁-C₄-alkyl (which is optionally substituted by fluorine and/or bromine), C₁-C₄-alkoxy (which is optionally substituted by fluorine and/or chlorine), represents C₁-C₄-alkylthio, C₁-C₄-alkylsulphinyl or C₁-C₄-alkylsulphonyl (which are optionally substituted by fluorine and/or chlorine), or represent di-(C₁-C₄-alkyl)-aminosulphonyl; where furthermore

R¹ represents the radical



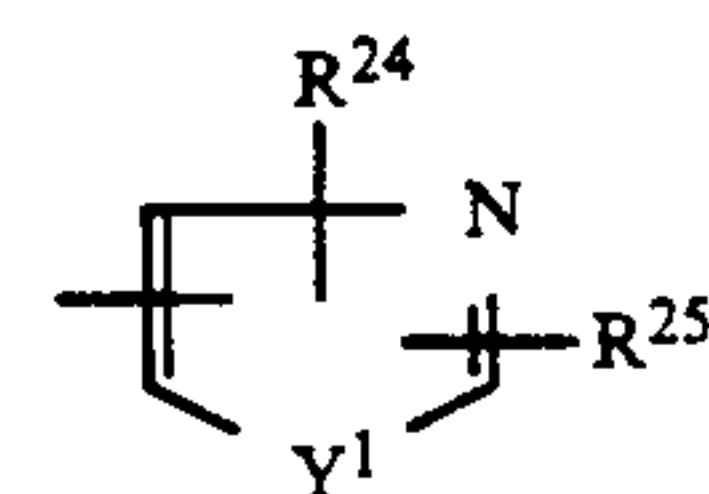
where

R²² and R²³ are identical or different and represent hydrogen, fluorine, chlorine, bromine, cyano, nitro, C₁-C₄-alkyl (which is optionally substituted by fluorine, chlorine, C₁-C₄-alkoxy and/or C₁-C₄-halogenalkoxy), C₁-C₄-alkoxy (which is optionally substituted by fluorine and/or chlorine), C₁-C₄-alkylthio, C₁-C₄-alkylsulphinyl or C₁-C₄-alkylsulphonyl (which is optionally substituted by fluorine and/or chlorine), di-(C₁-C₄-alkyl)-aminosulphonyl or C₁-C₄-alkoxycarbonyl, and

A represents oxygen, sulphur or the group N-Z¹, where

Z¹ represents hydrogen, C₁-C₄-alkyl (which is optionally substituted by fluorine, chlorine, bromine or cyano), C₃-C₆-cycloalkyl, benzyl, phenyl (which is optionally substituted by fluorine, chlorine, bromine or nitro), C₁-C₄-alkylcarbonyl, C₁-C₄-alkoxy-carbonyl or di-(C₁-C₄-alkyl)-aminocarbonyl; where furthermore

R¹ represents the radical

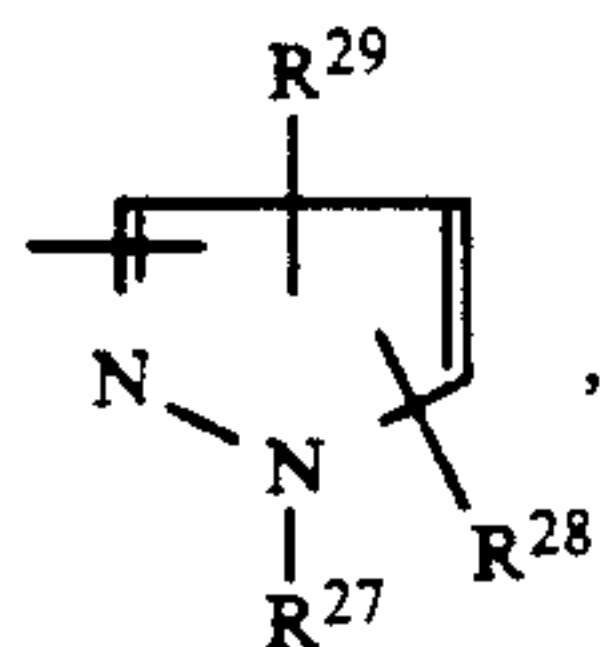


where

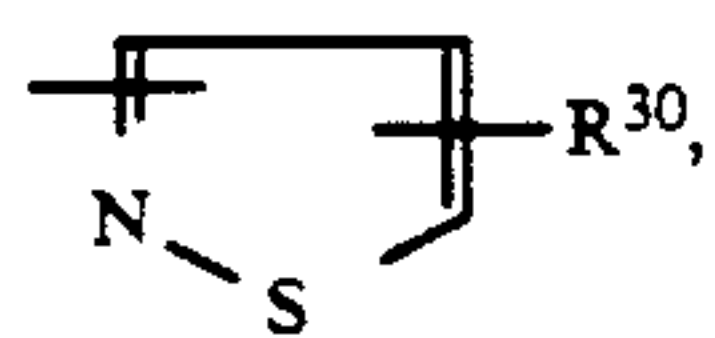
R²⁴ and R²⁵ are identical or different and represent hydrogen, C₁-C₄-alkyl, halogen, C₁-C₄-alkox-

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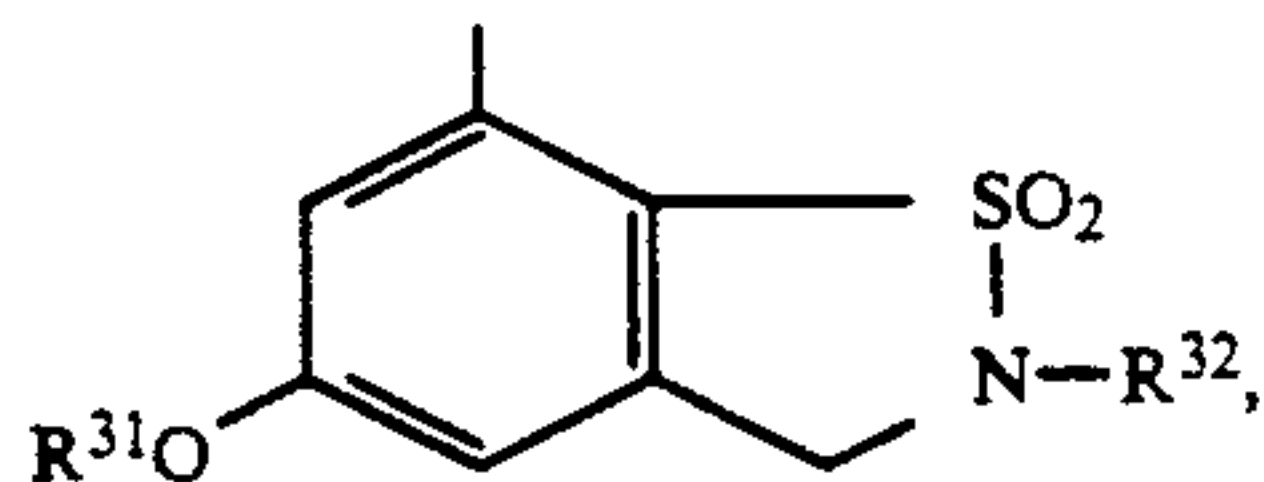
ycarbonyl, C₁-C₄-alkoxy or C₁-C₄-halogenalkoxy,
 Y¹ represents sulphur or the group N—R²⁶, where
 R²⁶ represents hydrogen or C₁-C₄-alkyl; where
 furthermore
 R¹ represents the radical



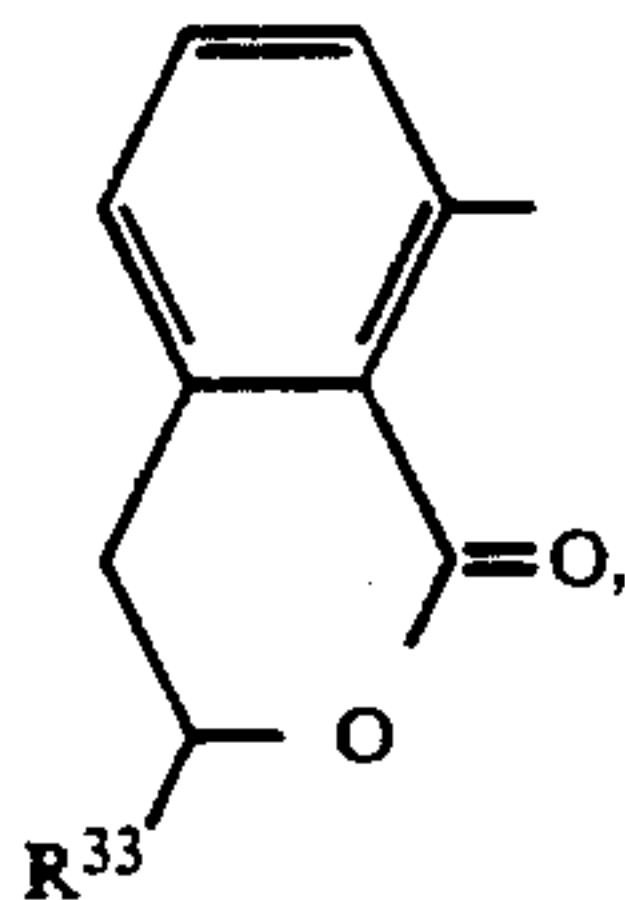
where
 R²⁷ represents hydrogen, C₁-C₄-alkyl, benzyl,
 (iso)quinolinyl or phenyl,
 R²⁸ represents hydrogen, halogen, cyano, nitro,
 C₁-C₄-alkyl (which is optionally substituted by
 fluorine and/or chlorine), C₁-C₄-alkoxy (which
 is optionally substituted by fluorine and/or chlo-
 rine), dioxolanyl or C₁-C₄-alkoxy-carbonyl and
 R²⁹ represents hydrogen, halogen or C₁-C₄-alkyl;
 where furthermore
 R¹ represents the radical



where
 R³⁰ represents hydrogen, halogen, C₁-C₄-alkyl,
 C₁-C₄-halogenalkyl, C₁-C₄-alkoxy, C₁-C₄-
 halogenalkoxy or C₁-C₄-alkoxy-carbonyl;
 where furthermore
 R¹ represents the radical



where
 R³¹ represents C₁-C₄-alkyl and
 R³² represents C₁-C₄-alkyl; where furthermore
 R¹ represents the radical



where — R³³ represents hydrogen or methyl
 in which furthermore
 R² represents hydrogen or the group —SO₂—R¹,
 in which furthermore
 R³ represents hydrogen, fluorine, chlorine, bromine,
 iodine, hydroxyl, mercapto, amino or an optionally
 fluorine- and/or chlorine-substituted radical selected
 from the group consisting of C₁-C₄-alkyl,
 C₃-C₆-cycloalkyl, benzyl, phenyl, C₁-C₄-alkoxy,

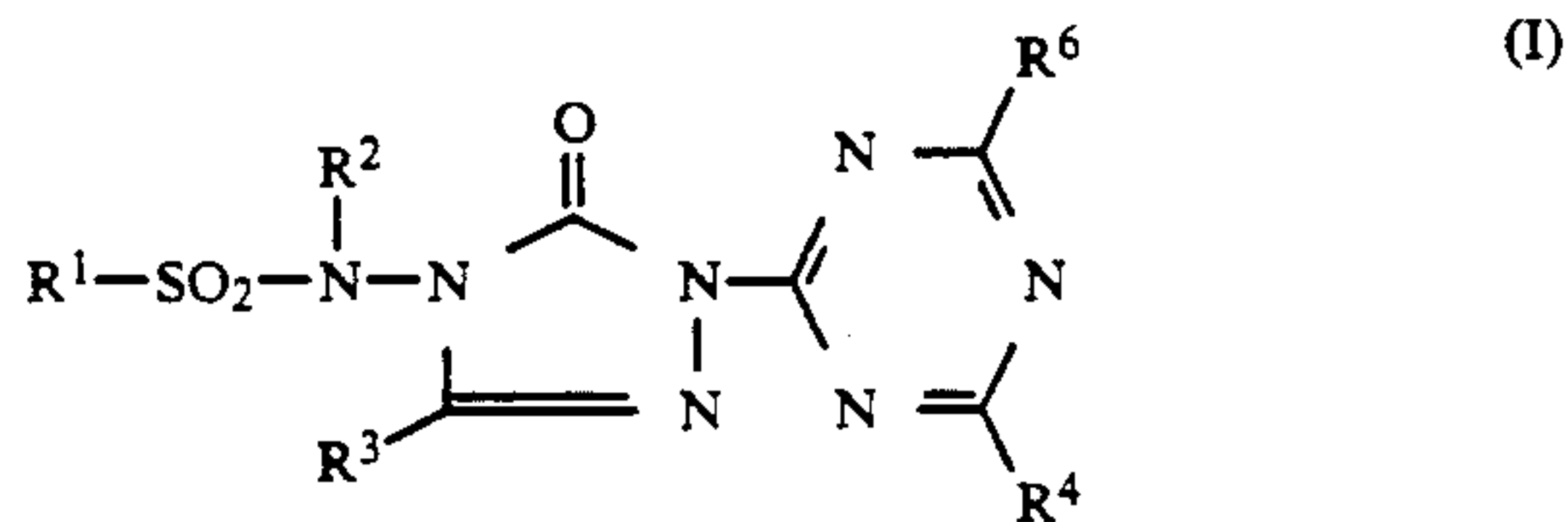
82

C₃-C₄-alkenyloxy, C₃-C₄-alkinyloxy, C₁-C₄-
 alkylthio, C₁-C₄-alkylsulphinyl, C₁-C₄-alkylsul-
 phonyl, C₃-C₄-alkenylthio, C₃-C₄-alkinylthio,
 benzyloxy, benzylthio, C₁-C₄-alkylamino and
 di-(C₁-C₄-alkyl)-amino,

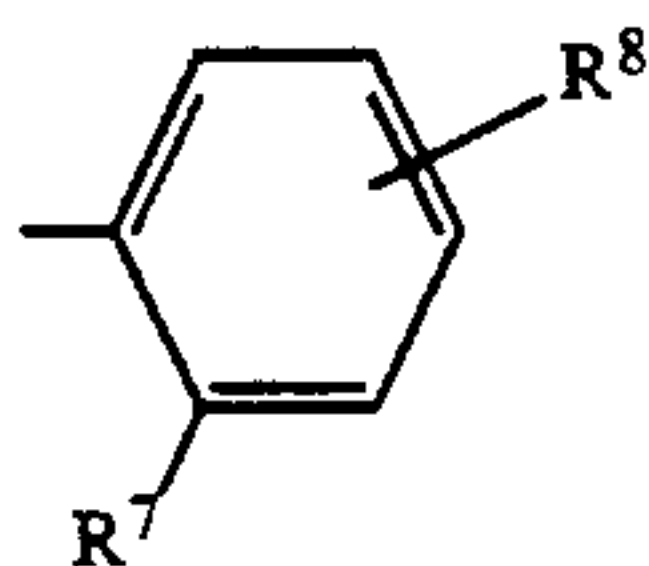
R⁴ represents hydrogen, fluorine, chlorine, bromine,
 hydroxyl, amino, C₁-C₄-alkyl, C₁-C₄-halogenoalk-
 kyl, C₁-C₂-alkoxy-C₁-C₂-alkyl, C₁-C₄-alkoxy,
 C₁-C₄-halogenoalkoxy, C₁-C₂-alkoxy-C₁-C₂-
 alkoxy, C₁-C₄-alkylthio, C₁-C₄-halogenalkylthio,
 C₁-C₄-alkylsulphinyl, C₁-C₄-alkylsulphonyl,
 C₁-C₄-alkylamino or di-(C₁-C₂-alkyl)-amino, and
 R⁶ represents hydrogen, fluorine, chlorine, bromine,
 C₁-C₄-alkyl, C₁-C₄-alkoxy, C₁-C₄-halogenoalk-
 oxy, C₁-C₄-alkylthio, C₁-C₄-alkylsulphinyl,
 C₁-C₄-alkylsulphonyl, C₁-C₄-alkylamino, dimeth-
 yl-amino or diethyl-amino,

or a salt thereof.

2. A compound or salt thereof of the formula



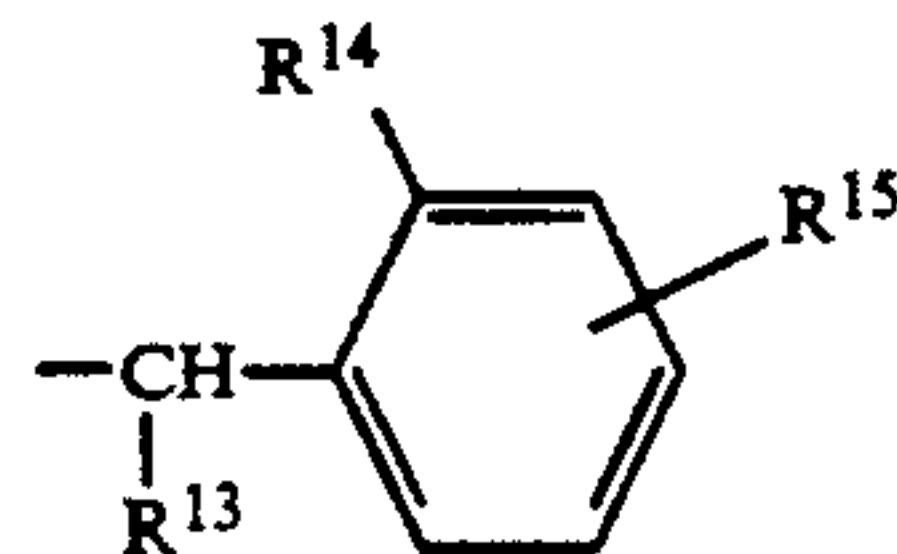
in which
 R¹ represents the radical



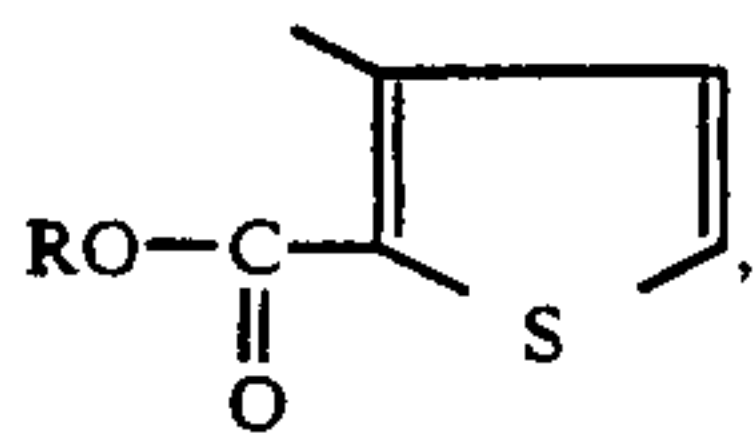
where
 R⁷ represents fluorine, chlorine, bromine, methyl,
 trifluoromethyl, methoxy, difluoromethoxy, tri-
 fluoromethoxy, C₁-C₃-alkylthio, C₁-C₃-alkyl-
 sulphinyl, C₁-C₃-alkylsulphonyl, dime-
 thylaminosulphonyl, N-methoxy-N-
 methylaminosulphonyl, phenyl, phenoxy or
 C₁-C₃-alkoxycarbonyl and

R⁸ represents hydrogen, fluorine or chlorine;
 where furthermore

R¹ represents the radical



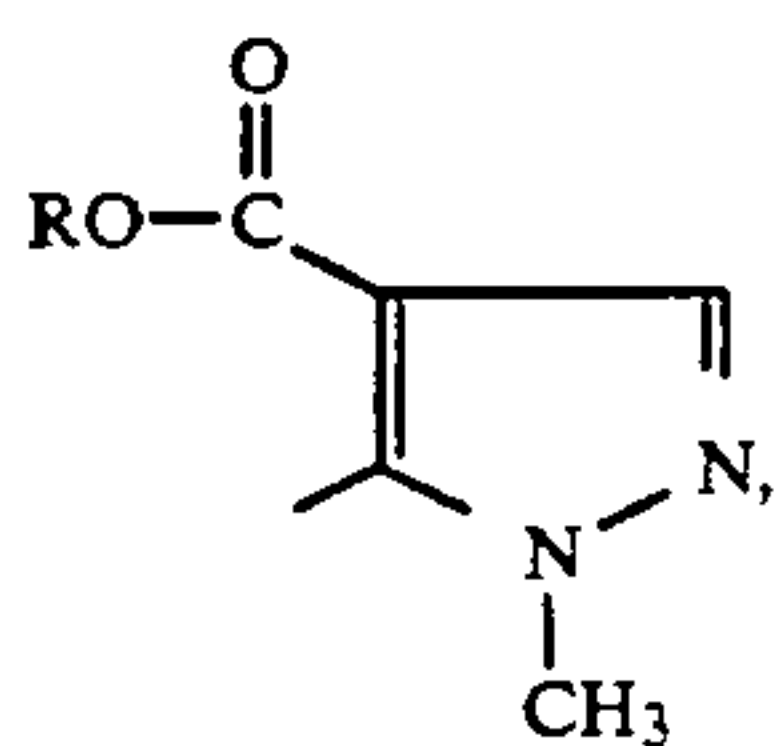
where
 R¹³ represents hydrogen,
 R¹⁴ represents fluorine, chlorine, bromine, methyl,
 methoxy, difluoromethoxy, trifluoromethoxy,
 ethoxy, methoxycarbonyl, ethoxycarbonyl, me-
 thylsulphonyl or dimethylaminosulphonyl and
 R¹⁵ represents hydrogen; where furthermore
 R¹ represents the radical



where

R represents C₁-C₄-alkyl, or

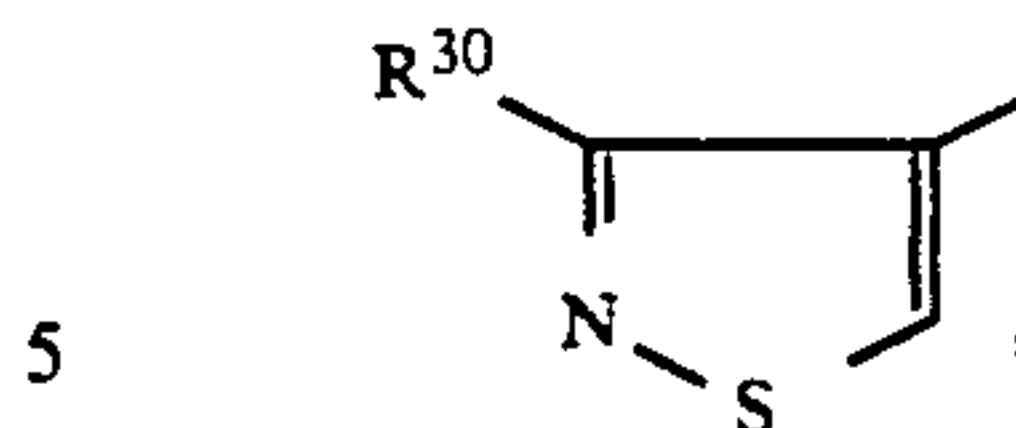
R¹ represents the radical



where

R represents C₁-C₄-alkyl, or

R¹ represents the radical



where

R³⁰ represents the hydrogen, chlorine, methyl, ethyl, methoxycarbonyl or ethoxycarbonyl;

in which furthermore

R² represents hydrogen or for the group —SO₂—R¹,

R³ represents hydrogen, methyl, ethyl, propyl, isopropyl, trifluoromethyl, cyclopropyl, benzyl, phenyl, t-butyl, s-butyl, i-butyl, n-butyl, methoxy or methylthio,

R⁴ represents hydrogen, fluorine, chlorine, bromine, methyl, trifluoromethyl, methoxy, ethoxy, difluoromethoxy, methylthio, ethylthio, amino, methylamino, ethylamino, dimethylamino or diethylamino, and

R⁶ represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl, methoxy, ethoxy, propoxy, isopropoxy, difluoromethoxy, methylthio, ethylthio, methylamino, ethylamino, dimethylamino or diethylamino.

3. A herbicidal composition comprising a herbicidally effective amount of a compound or salt thereof according to claim 1 or 2 and an inert diluent.

4. A method of combating unwanted vegetation which comprises applying to such vegetation or to a locus from which it is desired to exclude such vegetation a herbicidally effective amount of a compound or salt thereof according to claim 1 or 2.

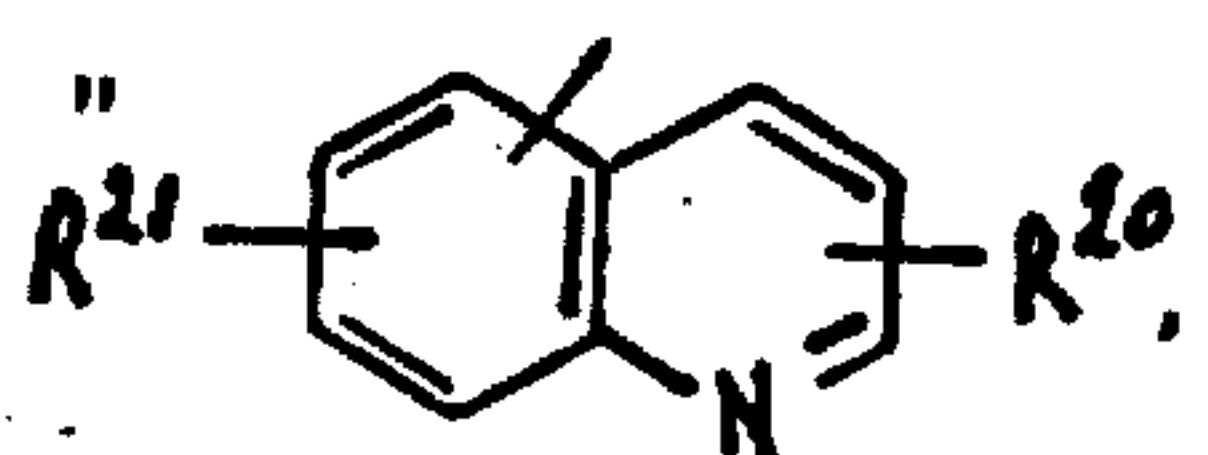
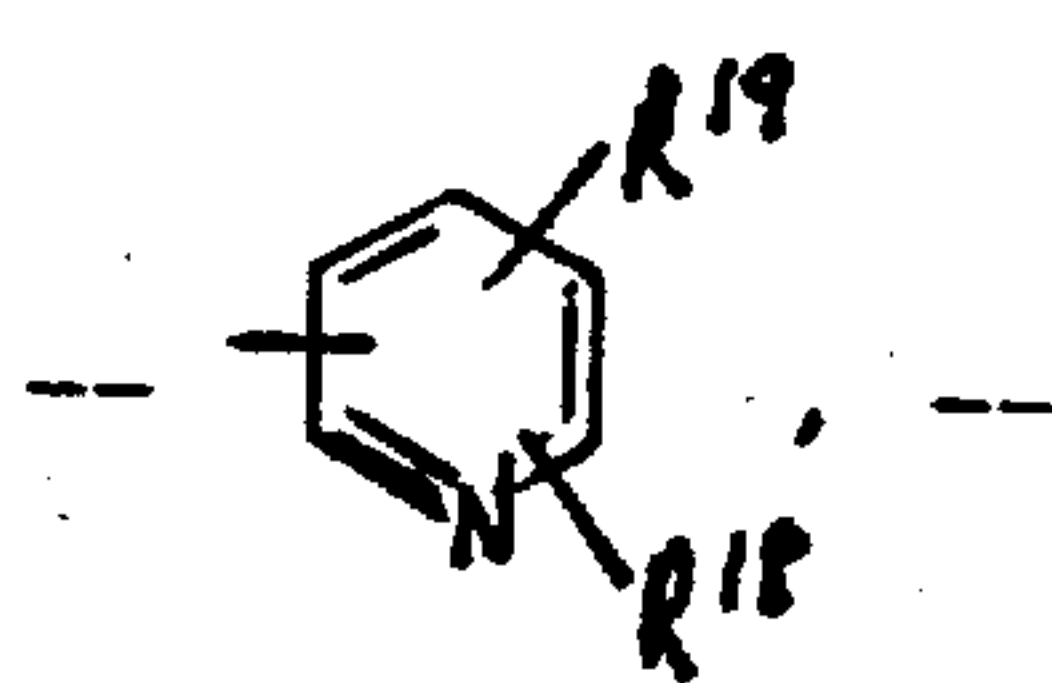
* * * * *

**UNITED STATES PATENT AND TRADEMARK OFFICE
CERTIFICATE OF CORRECTION**

PATENT NO. : 5,085,687
 DATED : February 4, 1992
 INVENTOR(S) : Muller et al.

Page 1 of 2

It is certified that error appears in the above-identified patent and that said Letters Patent is hereby corrected as shown below:

- | | |
|--------------------|--|
| Title Page | [75] Inventors: After " Klaus- " delete " Helmet " and substitute -- Helmut -- |
| Col. 77, line 52 | Delete " C ₁ -C ₆ -alkoxy " and substitute -- C ₁ -C ₆ -alkyl -- |
| Col. 78, line 24 | Delete " NOR ¹⁰ " and substitute -- NHOR ¹⁰ -- |
| Col. 79, line 58 | Delete "  " and substitute
 |
| Col. 80, line 40 | Delete " halogenalkoxy " and substitute -- halogeno-alkoxy -- |
| Col. 81, lines 1-2 | Delete " halogenalkoxy " and substitute -- halogeno-alkoxy |
| Col. 81, line 34 | Delete " halogenalkyl " and substitute -- halogeno-alkyl -- |
| Col. 81, line 35 | Delete " halogenalkoxy " and substitute -- halogeno-alkoxy -- |
| Col. 82, line 3 | Delete " C ₃ !4 " and substitute -- C ₃ - -- |

UNITED STATES PATENT AND TRADEMARK OFFICE
CERTIFICATE OF CORRECTION

PATENT NO. : 5,085,687

Page 2 of 2

DATED : February 4, 1992

INVENTOR(S) : Muller et al.

It is certified that error appears in the above-identified patent and that said Letters Patent is hereby corrected as shown below:

Col. 82, line 4 Delete " C₁!4 " and substitute -- C₁- --

Col. 82, line 10 Delete " halogenalkylthio " and substitute -- halogeno-alkylthio --

Signed and Sealed this

Twenty-fifth Day of January, 1994

Attest:



BRUCE LEHMAN

Attesting Officer

Commissioner of Patents and Trademarks